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RADIOLOGICAL SURVEYS OF IDAHO PHOSPHATE ORE PROCESSING—THE WET PROCESS PLANT

APRIL 1978

U.S. ENVIRONMENTAL PROTECTION AGENCY
OFFICE OF RADIATION PROGRAMS
LAS VEGAS FACILITY
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DISCLAIMER

This report has been reviewed by the Office of Radiation Programs - Las Vegas Facility, U. S. Environmental Protection Agency, and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for their use.

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PREFACE

The Office of Radiation Programs of the U.S. Environmental Protection Agency carries out a national program designed to evaluate population exposure to ionizing and nonionizing radiation, and to promote development of controls necessary to protect the public health and safety. This report describes various surveys which were conducted at the J. R. Simplot's Wet Process Plant near Pocatello, Idaho to provide basic data to be used for the evaluation of the radiological impact associated with the phosphate industry. Readers of this report are encouraged to inform the Office of Radiation Programs of any omissions or errors. Comments or requests for further information are also invited.

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ABSTRACT

Radiological surveys conducted at the J. R. Simplot's Wet Process Plant in Pocatello, Idaho indicate slightly elevated ambient levels of natural radioactivity. Compared to an estimated natural background annual dose equivalent rate of about 79 mrem, net gamma dose rates ranged from 42 mrem in general plant areas to 152 mrem per work year (2000 hours) on the ore piles. Ambient radon-222 concentrations, ranging from 0.14 to 1.9 pCi/l, were measured in various indoor locations. Elevated airborne radioactivity concentrations were measured in several work areas, with polonium-210 and radium-226 being the most predominant radionuclides of the natural uranium decay series. Particle size characterization indicates roughly 52 percent of the arithmetic total radioactivity is associated with the particle size fraction less than one micrometer equivalent aerodynamic diameter. Stack sampling results also show that appreciable concentrations of the naturally-occurring radionuclides are being discharged into the local environs. In general, the dose estimates and the interpretation of results have been oriented toward evaluating the maximum potential impact of the plant on the environment; however, no attempt has been made to determine the annual average dose to workers within the plant from all exposure pathways.

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INTRODUCTION

Recent reports (Windham et al., 1976; Guimond and Windham, 1975) indicate that mining and processing of phosphate ores technologically enhance the quantities of naturally-occurring radioactive materials such as radium and uranium available to man in the environment. Such radionuclides have relatively long half-lives and will therefore persist in man's habitat for thousands of years.

Current estimates show that the phosphate industry has generated about twenty times the volume of waste materials as compared to that from the uranium mining and milling industry. The total radioactive waste inventory generated by the uranium and the phosphate mining and milling industries are comparable to each other although the per gram radioactivity content of the phosphate waste is much less than that of uranium waste material (EPA, February 1977).

In general, the phosphate industry is neither regulated nor monitored for the possession, use, or discharge of radioactive materials associated with phosphate rock and its products and by-products. Recently, the State of Idaho (June 1, 1977) has prohibited the use of phosphate slag material in the construction of habitable structures, but has permitted the continued use of slag for road construction, railroad ballast, and other general purposes.

Of immediate concern is the accumulation of a data base which will ultimately lead to an assessment of the impact on public health due to the phosphate industry's activities. This report discusses various radiological surveys conducted in the J. R. Simplot's Wet Process Plant in Pocatello, Idaho. The intent of the study was to obtain a general indication of the concentrations of radionuclides in the ore, process streams, products, by-products, and effluents of the plant. In general, the dose estimates and the interpretation of results have been oriented towards evaluating the maximum potential impact of the plant on the environment. A similar study, conducted in a thermal process plant, has been discussed in another report.

THE WET PROCESS

Phosphate rock is shipped by rail from the Conda and Gay Mines to the J. R. Simplot's Wet Process Plant near Pocatello, Idaho. The rock is conveyed from the stockpile to the Calciner where it is heated to about 1500° F, burning out the organic material. This calcining also upgrades the phosph te content by two to three percent, with the calcined ore containing approximately 32 to 34 percent phosphorus pentoxide (P_2O_5). The calcined ore is ground into a fine powder which is then reacted with sulfuric acid in the Phosphoric Acid Plant to produce phosphoric acid and by-product gypsum. This reaction is approximated by the equation:

$$2\ H_2O + Ca_3(PO_4)_2 + 3\ H_2SO_4 \rightarrow 2\ H_3PO_4 + 3\ CaSO_4 \cdot 2\ H_2O$$

Water + Phosphate Ore + Sulfuric Acid → Phosphoric Acid + Gypsum

The phosphoric acid produced in this process is 30 to 32 percent P_2O_5. This acid is then used to make several grades of fertilizers. Phosphoric acid, ammonia and sulfuric acid are mixed together to produce various grades of ammonium phosphate and to make ammonium sulfate. Three basic grades of solid ammonium phosphate (ammo-phos), two grades of liquid ammo-phos, and crystalline ammonium sulfate are produced at the Wet Process Plant. These are 16-20-0, 18-46-0, and 11-54-0 for the solid ammo-phos; 10-34-0 and 11-37-0 for the liquid ammo-phos; and 21-0-0 for the ammonium sulfate. (The numbers represent the percent nitrogen, P_2O_5 and potash in the final product.)

In the manufacture of the solid ammo-phos, the ammonia, phosphoric acid, and sulfuric acid are mixed in a reactor to form a slurry. This slurry is then mixed with recycle fine product in

a granulator, with the slurry coating the outside of the recycle particles to form a layer of fresh ammo-phos. The granulated material is then dried and screened. The oversized material is crushed and recycled with the fine. The intermediate fraction, the ammo-phos product, is cooled and conveyed to storage.

The liquid fertilizer (10-34-0 and 11-37-0) is made by reacting ammonia, water, and concentrated "superacid" (70 percent P_2O_5) to form the liquid products. (The superacid is produced by evaporation of 52 percent P_2O_5.)

Ammonium sulfate is produced by reacting ammonia and sulfuric acid in a crystallizer, under vacuum, which forms a crystalline product. The crystals are separated from the liquid phase by centrifuging. The crystals are then dried and conveyed to storage and the liquid phase is returned to the crystallizer as a "seed" solution.

Another product, triple superphosphate (0-45-0) was produced by reacting the ground phosphate rock with phosphoric acid. This acidulated material is "cured" for a period of about three to four weeks and is then ground, granulated, dried, and screened. This process is no longer utilized; instead, a patented process is now used which eliminates the need for "curing." These various processes are shown in Figure 1.

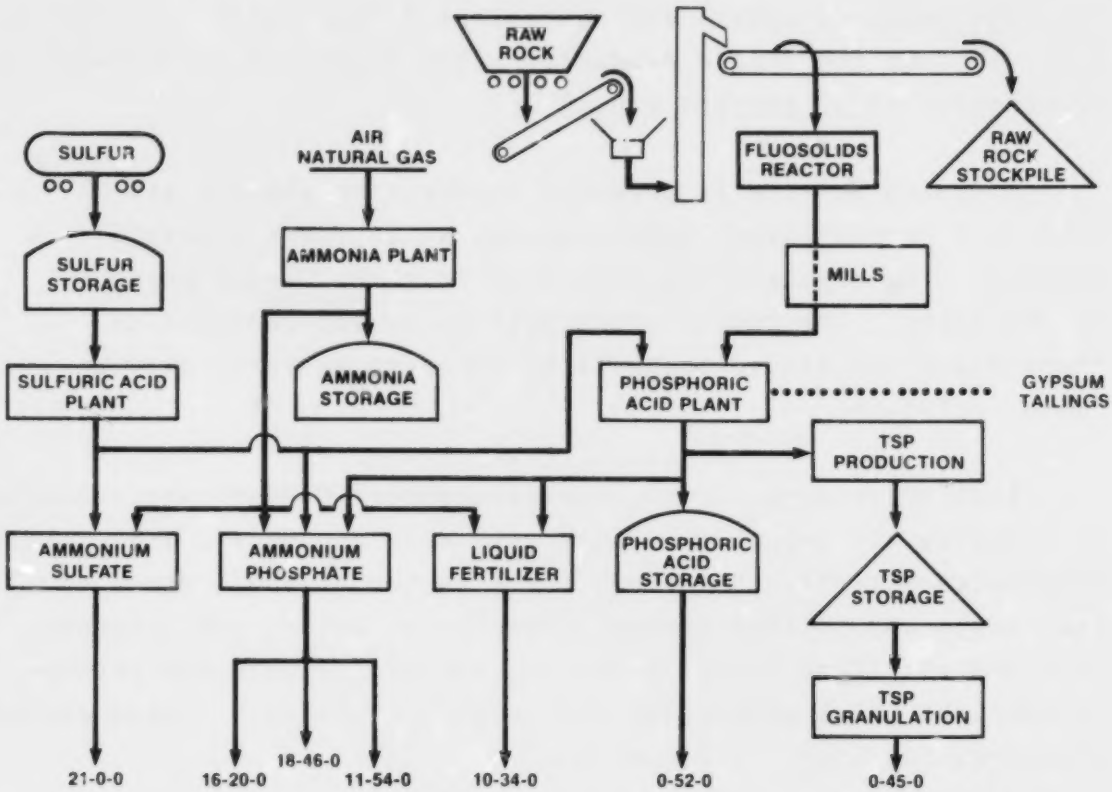

Figure 1. Flow diagram of the wet process

THE RADIOLOGICAL ASSESSMENT OF THE WET PROCESS PLANT

Radiological surveys were completed at the J. R. Simplot's Wet Process Plant in Pocatello, Idaho during the week of April 28 to May 1, 1975. Included among these surveys were: gamma radiation measurements; ambient radon-222 concentrations; radiological analyses of selected input, process and product samples; airborne particulate sampling; and particle-size characterization. Stack sampling of representative effluent discharge vents was also completed in September 1975. The following sections of this report discuss the results of these surveys.

GAMMA RADIATION SURVEYS

In order to assess the external gamma radiation exposure rates in the various working areas of the Wet Process Plant, a survey was conducted using a portable gamma scintillator survey meter.* This instrument was calibrated with a radium-226 standard, and measured the relative gamma radiation exposure rate in units of microroentgens per hour (μR/h). Measurements were made at a height of three feet above an area surface and the results shown in Table 1 represent average values for each location. At the time of these surveys, a pressurized ionization chamber (PIC) was not available to complete radiation surveys for comparison to the scintillator measurements. Although the scintillator survey meter's response is dependent on the energy of the gamma photon (Eadie et al., 1976), no instrument response correction factor has been applied to the reported scintillator measurements.

* Baird-Atomic, Type NE148A-Gamma Scintillator Ratemeter.

TABLE 1. GAMMA RADIATION SURVEYS[+]
GENERAL WORK AREAS

LOCATION DESCRIPTION	GROSS GAMMA EXPOSURE RATE μR/h	NET GAMMA EXPOSURE RATE* μR/h	WORK YEAR DOSE EQUIVALENT** mrem/year
200 Phosphoric Acid Plant:			
Below 24B filter	30	21	42
Evaporator Control Room	9	BKG.	BKG.
Ground floor, near front gate	30	21	42
Condensate pipe	120	111	222
Scrubber water	100	91	182
44-46% phosphoric acid tank, ground floor	30	21	42
48% phosphoric acid tank, acidulation	30	21	42
300 Phosphoric Acid Plant:			
Directly over 24C filter	70	61	122
Above 52% phosphoric acid storage tanks	16	7	14
Phosphoric acid control room	9	BKG.	BKG.
27-29% Phosphoric acid tank, outside 200/300 phosphoric acid plants	14	5	10
42% Phosphoric acid tank, next to north rotating filter	12	3	6
70% Phosphoric acid tanks	8	BKG.	BKG.
Storage Areas:			
Ammonium sulfate	8	BKG.	BKG.
Ammonium phosphate (18-46-0)	10	1	2
Ammonium phosphate (16-20-0)	10	1	2
Ammonium phosphate (11-54-0)	11	2	4
Roadway near (11-54-0) storage area	30	21	42
Sulfur	30	21	42

TABLE 1. GAMMA RADIATION SURVEYS+
GENERAL WORK AREAS (Cont'd)

LOCATION DESCRIPTION	GROSS GAMMA EXPOSURE RATE μR/h	NET GAMMA EXPOSURE RATE* μR/h	WORK YEAR DOSE EQUIVALENT** mrem/year
Triple super phosphate green pile	35	26	52
Triple super phosphate (0-45-0), final product pile	30	21	42
Ore Pile:			
Gay mine, high grade	70	61	122
Conda mine, high grade	85	76	152
Conda mine, low grade	70	61	122
West Plant Outfall, discharge to river	20	11	22
West Plant Outfall, nearby ground	14	5	10
Cold Pit Area	35	26	52
Mill Building (Grinder)	45	36	72
Urea tank area, asphalt	45	36	72
Urea tank area, nearby ground	15	6	12
New gypsum pile, underflow pond area	25	16	32
New gypsum pile, center	40	31	62
Gypsum standing water (end of pile)	20	11	22
Thermal plant slag	60	51	102
Background Radiation			
General Pocatello, Idaho area	9	--	18 mrem per Work Year 79 mrem per Year for continuous exposure

+These surveys were conducted on April 30, 1975 using the Baird-Atomic NE148A-Gamma Scintillator Ratemeter. All measurements are average instrument response, in units of μR/h, at three-foot height above surface area.

*An estimated background gamma exposure rate of 9 μR/h (using the scintillator survey meter) has been subtracted from the gross value to obtain the net result. (This instrument does not respond fully to cosmic radiation. Thus, neither the estimated background nor the gross readings reflect the true contribution of the cosmic component.)

**Work Year has been defined as 40 h/wk for 50 wk/y (i.e., 2000 hours). The Work Year Dose Equivalent is the net value multiplied by the Work Year time equivalent; i.e., the dose estimate due to continuous work year occupancy per specific area. This may be an overestimate, depending on the actual occupancy factor.

Therefore, the reported scintillator measurements (Table 1) are assumed to be equivalent to the "true" gamma exposure rate due to both the cosmic and the terrestrial components.*

EXTERNAL DOSE ASSESSMENT

Background Radiation

Gamma exposure rate data for non-coastal plain regions of the United States have been summarized and indicate a mean value of 5.2 μrem/h (46 mrem/year) absorbed dose rate equivalent in air from terrestrial sources (NCRP, November 1975). Specific surveys around the National Reactor Testing Station near Idaho Falls, Idaho showed an average terrestrial dose equivalent rate of about 6 μrem/h (Oakley, 1972). Therefore, the natural terrestrial background radiation annual dose equivalent for the Pocatello, Idaho area should be approximately 53 mrem per year (i.e., continuous exposure at 6 μrem/h x 24 h/day x 365 day/year). The scintillator instrument's response to cosmic radiation in the Pocatello area has been estimated to be roughly 3 μR/h. This value corresponds to an annual average cosmic dose equivalent rate of 28 mrem per year as summarized by NCRP (November 1975). Therefore, a total natural background radiation exposure rate of 9 μR/h has been used to obtain the "net" exposure rates for the survey results as reported in Table 1.

In-Plant Radiation Exposures

Several areas of background radiation exposure rates were measured in the Wet Process Plant (Table 1). The lowest net exposure rate (measured minus background rate) was 1 μR/h in the

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* The absorbed exposure rates (mR/year) have been converted to dose equivalents (mrem/year), assuming the roentgen is essentially equal to a rad and using a quality factor of one for gamma photons. This is consistent with the recommendations of the National Council on Radiation Protection (NCRP, January 1971).

ammonium sulfate storage area and in the area of the 70 percent phosphoric acid tanks. Therefore, for workers who spend the majority of their working periods in these areas, the maximum annual dose equivalent would be about 2 mrem per year (i.e., 1 μR/h x 40 h/wk x 50 wk/year). General working areas within the 200 Phosphoric Acid Plant had net exposure rates ranging from background in the evaporator control room to 21 μR/h around the phosphoric acid tanks. This corresponds to a maximum dose equivalent of 42 mrem per year. The highest net exposure rate, 111 μR/h, obtained in the Wet Process Plant was observed near a condensate pipe in the 200 Phosphoric Acid Plant.

Background exposure rates were also measured in the phosphoric acid control room of the 300 Phosphoric Acid Plant. Area surveys over the ore storage piles showed net exposure rates of about 61 μR/h for the low grade ores to 76 μR/h for the high grade ore. Surveys around the various plant product areas ranged from background in the ammonium sulfate storage area to a high of 26 μR/h on the triple super phosphate (green pile) storage area.

In-plant radiation exposures are also summarized in Table 1. By assuming continuous exposure during the entire work period (40 h/wk x 50 wk/year) at the net gamma exposure rate levels, the reported dose equivalents are probably overestimated. A more precise dose estimate could be obtained by fractioning the time spent in various work locations and using that particular exposure rate; however, such information has not been obtained in this study. It should also be emphasized that these in-plant external gamma dose estimates for the work year are in addition to the dose received by an individual due to natural background radiation exposure. Furthermore, these estimates are based on free air measurements. It is estimated that the dose to internal body organs varies from 0.5 to about 0.7 of the free air measurement due to the shielding effect of the body (O'Brien and Sanna, 1976).

Surveys conducted at a Thermal Process Plant in Florida (Windham et al., 1976) showed external gamma dose rates slightly higher than those measured in this study. Total measured dose equivalent rates (including background) in the Florida plant ranged from 100 mrem per work year in general plant areas to a maximum of 300 mrem per work year in the ferrophosphorus and slag storage area. For the Idaho Thermal Process Plant, the general work areas averaged about 42 mrem per work year, and the maximum net dose equivalent rate (on the slag pile) was 182 mrem per work year, above a background level of about 79 mrem per year. For the Idaho Wet Process Plant, the general work area averaged about 42 mrem per work year, and the maximum net dose equivalent rate on the ore piles was 152 mrem per work year, above a background level of about 79 mrem per year.

AMBIENT RADON-222 CONCENTRATIONS

Sampling System and Analytical Methods

A continuous, low-volume sampling system was used to obtain samples of ambient air for analysis of radon content (U.S. Public Health Service, 1969). This sampling technique consists of drawing filtered air through a small, low-volume air pump (less than 10 ml/min sampling rate) into a 30-liter Mylar bag. The air intake was about one meter above the ground surface. Various sampling time periods were used for this study.

Radon analysis was completed at the Environmental Monitoring and Support Laboratory in Las Vegas, Nevada (EMSL-LV) using a radon concentration apparatus for sample preparation (Johns, 1975). This apparatus permits the isolation of radon from about a 5-liter ambient air sample by transferring the ambient air from the Mylar bag into a container of known volume, followed by circulation through water and carbon dioxide traps. Radon is collected on two charcoal traps maintained at a temperature of about minus 80° C using dry ice and acetone. The radon sample is

then de-emanated into a Lucas scintillation cell (125-ml volume) using helium gas at 400° C. The Lucas cell is held for four and one-half hours to allow for the ingrowth of the radon daughters and then counted in a photomultiplier tube/scaler unit. The radon activity is calculated to the mid-point of the sample collection period to account for the radioactive decay ($T_{\frac{1}{2}}$ = 3.82 days) of radon from the time of sample collection to time of analysis. The ambient air volume sampled is determined by correcting for air density differences based on the nearest 1000-foot elevation increment, and the average ambient temperature during the sampling period. The radon concentrations (in pCi/l) reported in Table 2 therefore represent the average ambient radon concentration for the sampling period for each specific sampling site location.

Radon Measurement Results

Ambient radon-222 concentrations measured in the various indoor working areas ranged from a low of 0.14 pCi/l near a dryer of the No. 100 Ammophos Plant to a high of 1.9 pCi/l outside the control room of the 300 Phosphoric Acid Plant. Out-of-doors, the ambient radon ranged from 0.23 pCi/l to 0.31 pCi/l on the ore loading and unloading areas and the gypsum pile, respectively.

Harley (1975) estimated the average background concentration of radon in surface air of about 0.10 pCi/l in the northern hemisphere. Other authors (Pearson, 1967; United Nations, 1972) have reported radon concentrations in the general environment ranging from 0.03 to 0.4 pCi/l. Ambient outdoor radon concentrations measured in areas associated with inactive uranium mill tailings piles indicate average levels of 0.34 pCi/l (Monticello, Utah), 0.38 pCi/l (Salt Lake City, Utah), 0.51 pCi/l (Durango, Colorado), and 0.83 pCi/l (Grand Junction, Colorado), (U.S. Public Health Service, 1969).

Indoors, the radon levels are typically from three to four times the outdoor levels due to the entrapment within the structure of radon emanation from soils beneath the structure and also from radon emanated from the building materials, supplemented by radon infiltration from outside air. In general, radon levels inside structures are usually 0.6 to 1.2 pCi/l for ventilation rates below four air changes per hour (Johnson et al., 1973). As shown in Table 2, the ambient radon concentrations measured in the Wet Process Plant are within expected ranges due to natural terrestrial background sources alone.

Radon Dosimetry

The report , Swift et al. (1976) discusses various aspects of radon dosimetry including the degree of equilibrium between

TABLE 2. AMBIENT RADON-222 CONCENTRATIONS

Date Time On	Off	Location Description	Radon Concentration * (pCi/l)
4/28/75 1155	1542	Calciner Bldg. No. 3 Calciner	0.18 ± 0.036
4/28/75 1035	1603	300 Phosphoric Acid Plant Outside Control Room	1.9 ± 0.12
4/29/75 0940	1612	100 Plant, Ground Floor Near Ammophos Dryer	0.14 ± 0.033
4/29/75 1021	1530	TSP Storage Area	0.55 ± 0.064
4/30/75 0904	1555	Grinder Building Mill #7	0.22 ± 0.041
4/30/75 0828	1500	TSP Dryer Station 13	0.32 ± 0.05
5/1/75 0930	1332	Gypsum Pile- North End	0.31 ± 0.049
5/1/75 0942	1320	Ore Loading and Unloading Area	0.23 ± 0.043

* Radon-222 concentration ± two-sigma counting error term. To convert to pCi/m^3, multiply the pCi/l value in the table by 1000.

radon and its daughters, the fraction of unattached daughter ions and certain assumptions regarding exposure modes to tissues within the lung. For dose estimates in this study, a dose conversion factor of 4 mrem per year, for continuous exposure to 1 pCi/m^3, was used. This factor represents radon-222:polonium-218:lead-214:bismuth-214 concentrations of 1.0:0.90:0.51:0.35 pCi/m^3, respectively. This is equivalent to an average daughter product equilibrium ratio of 50 percent and an average ventilation rate of one air change per hour, which is typical in living accommodations with adequate ventilation. This dose is delivered to the basal cell nuclei of segmental bronchi which are estimated to lie 60 μm below the surface where the activity is deposited. A quality factor of 10 for alpha particles has been used in the dose calculations. Therefore, continuous exposure to an assumed average background radon concentration of 100 pCi/m^3 (0.10 pCi/l as estimated by Harley, 1975) would result in a dose equivalent of 400 mrem per year. For a worker exposed for an entire work year (40 h/wk x 50 wk/year) to the maximum ambient measured radon concentration of 1,900 pCi/m^3, the dose equivalent would be about 1.7 rem per year.*

WET PROCESS SAMPLES

Appendix A discusses the radiochemical analytical techniques used for this study. Since the completion of the analyses of the following samples, a question has arisen as to the validity of the lead-210 radiochemical procedure. Although the procedure as outlined in Appendix A for the lead-210 analysis was fully tested and proved satisfactory for water sample analysis, there now appears to be some difficulty in obtaining consistent lead-210 recovery rates for solid samples such as the ore, briquettes,

* $4 \text{ mrem/year/(pCi/m}^3) \times \frac{2000 \text{ h Work Year}}{8760 \text{ h Continuous Exposure}} \times 1{,}900 \text{ pCi/m}^3 \times 10^{-3} \text{ rem/mrem} = 1.7 \text{ rem/year}$

The significance of this dose estimate is questionable, given the uncertainties in the determination of background and the "grab sample" nature of this particular measurement, plus the actual exposure conditions of the plant personnel.

slag, and air filter samples. The inaccuracies of the lead-210 results relate to analysis of samples without the benefit of a tracer or carrier yield. Thus the assumed yield, which was assumed to be the same as that for test samples (i.e., 85 percent) was in error by up to a factor of five. Comparison of lead-210 cross-check results, as reported for two EPA laboratories, indicates that the EMSL lead analyses are roughly a factor of five lower than the actual lead-210 content (see Appendix C for data which limits the uncertainty for individual results). This error has also been noted in recent cross-check air sample results. Since the majority of samples have been consumed to obtain the results reported here, it is impossible to re-analyze these samples to resolve this question. Therefore, the lead-210 results which have been obtained using the EMSL procedure (Appendix A) have been reported here, but for solid samples, these results probably underestimate the real sample lead-210 activity by up to about a factor of five. Future lead-210 analyses will be conducted with a revised technique.

The problem of underestimating the lead-210 content is also reflected in the estimate of a sample's polonium-210 content, as discussed in Appendix A. Results reported in the thermal process report (ORP/LVF, 1977) for selected samples (e.g. calcined ore and slag) indicate that the polonium-210 values reported here may overestimate the actual polonium-210 content by a factor of ten or more (ranging from 1 up to about 20, but could be higher); i.e., if the reported EMSL lead-210 results are actually a factor of five too low. Recognizing these difficulties encountered in obtaining valid lead-210 and polonium-210 results, the following sections describe the radiological evaluations of the Wet Process.

The two-sigma (95 percent) statistical counting error is reported for all of the radiochemistry data presented in this report. The techniques for calculating the counting errors are illustrated in Johns (1975). The error terms for data averages, the decay and ingrowth corrected polonium-210 results, and for the net air sample results represent error terms obtained using

techniques for propagating error estimates as given in Appendix A and by Eadie and Bernhardt (1976).

It should be recognized that the statistical counting error only includes the uncertainty from the number of counts measured from the sample and instrument background. It is not an estimate of the total analytical uncertainty nor uncertainties in sample collection, preparation and/or aliquoting procedures. Evaluation of soil sampling and a..alytical techniques for plutonium by Bernhardt (1976) indicate that sampling and analytical variations of up to 50 percent and more (95 percent confidence level) are observable in many groups of data, especially at levels within about an order of magnitude of the analytical detection level. Thus, it should be recognized that the uncertainty of the results is greater than the counting error.

Table 3 presents the results of analyses of samples considered to be typical of the raw materials input to the wet process. The average radium-226 content of the Idaho phosphate rock (ore) was 29 pCi/g, and the calcined ore contained 38 pCi/g for the Wet Process Plant, but was 26 and 25 pCi/g, respectively, for the Thermal Process Plant (ORP/LVF, 1977). Radium-226 analysis of Florida ore average about 60 pCi/g (Guimond and Windham, 1975).

The results of limited analyses of phosphoric acid samples are shown in Table 4. Radium-226 concentrations in the various grades of phosphoric acid ranged from 5.7 ± 0.87 to 63 ± 3.3 pCi/l.

The results of analyses of several gypsum samples are shown in Table 5. The radium-226 content of a composite sample from the gypsum pile was 23 ± 0.87 pCi/g. For the samples from the phosphoric acid reactor and the thickner, the suspended matter was allowed to "settle-out" of solution and hence a suspended and liquid fraction of the sample were analyzed. For both samples,

TABLE 3. RAW MATERIALS INPUT TO THE WET PROCESS* (SAMPLED 4/30/75)
RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

Radionuclide	Conda Low Grade Ore pCi/g	Conda Main Bed Ore pCi/g	Gay High Grade Ore pCi/g	Ore Calciner Product pCi/g	Raw Sulphur pCi/g	Sulphuric Acid 98% pCi/l	Urea pCi/l
Ra - 226	32 ± 1.0	28 ± 0.97	28 ± 0.96	38 ± 1.1	0.11 ± 0.065	0.52 ± 0.17	1.1 ± 0.44
Po - 210	48 ± 5.9	32 ± 3.4	35 ± 3.4	42 ± 6.2	< 0.32	NA	51 ± 19
U - 234	< 15	39 ± 30	28 ± 8.1	64 ± 5.0	< 0.091	NA	0.70 ± 0.10
U - 235	< 8.8	< 9.2	< 1.6	1.4 ± 0.38	< 0.047	NA	< 0.033
U - 238	18 ± 16	< 20	26 ± 7.5	38 ± 3.2	< 0.091	NA	0.67 ± 0.10
Th - 230	56 ± 4.1	45 ± 6.6	44 ± 7.7	65 ± 3.2	0.046 ± 0.029	NA	1.5 ± 0.23
Th - 232	1.9 ± 1.7	< 1.3	2.9 ± 2.3	0.40 ± 0.27	< 0.0077	NA	0.10 ± 0.067
Pb - 210	1.9 ± 0.62	12 ± 0.96	7.9 ± 0.84	9.0 ± 0.89	0.61 ± 0.56	< 0.90	13 ± 2.4
Ra - 228	< 0.86	< 0.95	1.1 ± 0.96	< 0.96	< 0.96	< 0.62	5.1 ± 3.6

* NA indicate no analysis.

TABLE 4. PHOSPHORIC ACID SAMPLES (SAMPLED 4/30/75)
RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM, in pCi/l

Radionuclide	Phosphoric Acid 70%	Phosphoric Acid 52%	Phosphoric Acid 48%	Phosphoric Acid 44-46%	Phosphoric Acid 42%	Phosphoric Acid 27-34%
Ra - 226	63 ± 3.3	6.7± 0.95	40 ± 2.3	8.5 ± 1.1	24 ± 1.8	5.7 ± 0.87
Pb - 210	250 ± 8.9	82 ± 5.0	41 ± 3.8	140 ± 6.1	95 ± 5.2	85 ± 4.9
Ra - 228	10 ± 4.8	5.9± 3.7	5.1 ± 3.7	3.9 ± 3.6	5.6 ± 3.7	< 4.1

TABLE 5. GYPSUM SAMPLES* (SAMPLED 4/30/75)
RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

Radionuclide	Gypsum Pile Composite (Solid) pCi/g	Gypsum Slurry from Reactor (Suspended) pCi/g	Gypsum Slurry from Reactor (Liquid) pCi/l	Gypsum Thickner Overflow- (Suspended) pCi/g	Gypsum Thickner Overflow- (Liquid) pCi/l	Gypsum Pond Water (Total) pCi/l	Gypsum Pond Water Overflow (Total) pCi/l	New Gypsum Pile Water Underflow (Total) pCi/l
Ra-226	23 ± 0.87	43 ± 1.2	15 ± 1.4	48 ± 1.8	39 ± 2.3	61 ± 2.9	40 ± 2.3	36 ± 2.2
Po-210	26 ± 3.0	5.7 ± 0.68	--	14 ± 1.5	<40	<5.4	ND	37 ± 1.1
U-234	24 ± 20	2.7 ± 0.32	--	3.3 ± 0.24	--	910 ± 66	270 ± 24	550 ± 44
U-235	<6.8	0.09 ± 0.04	--	0.15 ± 0.03	--	35 ± 3.8	9.1 ± 1.1	28 ± 2.4
U-238	<14	2.9 ± 0.32	--	3.3 ± 0.24	--	900 ± 66	270 ± 24	560 ± 44
Th-230	26 ± 7.1	5.7 ± 0.33	--	13 ± 0.40	0.24 ± 0.12	2.6 ± 1.0	77 ± 2.4	78 ± 2.0
Th-232	<1.3	0.08 ± 0.04	--	0.17 ± 0.05	<0.04	<0.20	0.80 ± 0.24	0.64 ± 0.16
Pb-210	1.1 ± 0.96	3.3 ± 0.41	--	4.3 ± 0.31	38 ± 1.9	68 ± 9.2	50 ± 2.6	100 ± 3.1
Ra-228	<1.0	1.7 ± 0.91	3.9 ± 3.6	<1.8	<3.4	<3.7	<3.7	<3.7

* ND indicates non-detectable; blanks indicate no data.

appreciably more radioactivity was contained in the suspended versus the liquid fraction, perhaps reflecting the water insolubility of these radionuclides. Total water samples, i.e., analyzed for both the liquid and suspended fractions, from the gypsum pond also show elevated concentrations of radioactivity.

The results of analyses of the various product fertilizers are shown in Table 6. The highest radium-226 activity content of 14 ± 0.68 pCi/g was measured in the triple superphosphate (TSP). The liquid fertilizer (10-34-0) had a radium-226 content of 25 ± 1.8 pCi/l.

ENVIRONMENTAL SAMPLES

Grab water samples were obtained from the Portneuf River at locations both upstream and downstream from the liquid waste effluent discharge from both the Thermal Process and the Wet Process Plants. Prior to analysis, the suspended solids were allowed to "settle-out" and hence, both a liquid (soluble component) and a suspended (insoluble component) fraction of each water sample was processed. The water samples collected during this study are non-potable, and human ingestion of such water is improbable.

The radiochemical results for the Portneuf River samples are shown in Table 7. For the liquid fraction, the results of the upstream and downstream samples are essentially identical since they are within the two-sigma counting error term for each respective analysis. Similar analytical agreement was not obtained from the results of the suspended fraction analyses. Although the measured radium-226 content was lower in the downstream sample, the uranium and thorium contents were about an order of magnitude greater in the downstream versus the upstream sample. The higher radioactivity content in the suspended fraction may indicate the insolubility of waste-product effluents. It must be emphasized here that these results are based on

TABLE 6. FERTILIZER SAMPLES (SAMPLED 4/30/75)
RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERMS

Radionuclide	Triple Super Phosphate (TSP) 0-45-0 Green pCi/g	Ammonium Sulphate 21-0-0 pCi/g	Ammonium Phosphate 11-54-0 pCi/g	Ammonium Phosphate 18-46-0 pCi/g	Ammonium Phosphate 16-20-0 pCi/g	Liquid Fertilizer 10-34-0 pCi/l
Ra - 226	14 ± 0.68	0.10 ± 0.071	0.85 ± 0.18	0.65 ± 0.16	1.6 ± 0.23	25 ± 1.8
Po - 210	18 ± 2.1	< 0.47	4.6 ± 0.70	2.1 ± 0.86	3.7 ± 0.69	45 ± 33
U - 234	45 ± 19	< 1.3	< 20	20 ± 3.5	< 19	140 ± 8.7
U - 235	< 3.3	< 0.84	< 11	0.76 ± 0.24	< 13	5.3 ± 0.58
U - 238	44 ± 18	< 1.2	27 ± 25	21 ± 3.5	< 24	140 ± 8.6
Th - 230	54 ± 8.6	< 0.16	63 ± 9.7	6.2 ± 0.32	29 ± 6.4	440 ± 5.1
Th - 232	< 1.6	< 0.11	2.0 ± 1.9	0.079 ± 0.038	< 1.4	3.8 ± 0.51
Pb - 210	3.6 ± 0.73	1.3 ± 1.1	0.62 ± 0.54	1.8 ± 0.6	1.2 ± 0.58	5.3 ± 2.2
Ra - 228	< 0.88	< 0.89	< 0.96	1.8 ± 1.0	< 0.92	11 ± 3.9

TABLE 7. PORTNEUF RIVER SAMPLES*
RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM.
LIQUID FRACTION IN UNITS OF pCi/l, AND SUSPENDED FRACTION IN UNITS OF pCi/g

Radionuclide	Upstream, East Bank - Liquid	Downstream, East Bank - Liquid	Upstream, East Bank-Suspended	Downstream, Eastbank-Suspended
Ra-226	0.70 ± 0.35	0.54 ± 0.33	3.6 ± 1.7	1.8 ± 1.3
Po-210	ND	ND	-	<2.4
U-234	1.2 ± 0.22	1.2 ± 0.20	0.067 ± 0.019	0.70 ± 0.20
U-235	<0.031	<0.029	<0.0095	<0.10
U-238	0.55 ± 0.15	0.51 ± 0.11	0.067 ± 0.019	0.70 ± 0.20
Th-230	<0.062	<0.029	0.17 ± 0.048	1.6 ± 0.40
Th-232	<0.031	<0.029	0.17 ± 0.048	1.7 ± 0.40
Pb-210	2.7 ± 1.3	<1.4	-	6.1 ± 1.6
Ra-228	<3.6	<4.0	<20	-

* Grab sampled 5/8/75.
ND indicated non-detectable; blanks indicate no data.

analysis of only one grab sample from both the upstream and the downstream flow. Also, the measured radioactivity concentrations of both the liquid and suspended fractions of both of these river water samples are well within the typical background range for naturally-occurring radioactivity (Holtzman, 1964). The slightly elevated activity in the suspended sediments indicate that any future efforts to resolve whether there is any contribution of radioactivity to the stream should include sediment sampling.

Additional water sample results are shown in Table 8. The incoming water (Well #5) and the discharge water from the West Plant Outfall are at typical background radioactivity concentrations. The cold pit water sample, however, shows an elevated radioactivity content of the suspended fraction, again reflecting the water insolubility of these natural radionuclides. The cold pit water is recycled water within the plant which is not discharged to the uncontrolled environment.

SCRUBBER DISCHARGE SAMPLES

Results of the suspended and liquid fractions of various scrubber discharge samples are presented in Table 9. Again, the suspended fraction contains the greater radioactivity content. The lowest radium-226 concentration of 2.5 ± 1.1 pCi/g was measured in the suspended fraction of the 200 Plant-phosphoric acid scrubber discharge sample. The highest radium-226 content of 33 ± 1.1 pCi/g was obtained from the suspended fraction of the calciner scrubber discharge. These results are similar to those obtained from the calciner scrubber discharges from the Thermal Process Plant (ORP/LVF, 1977).

AIRBORNE PARTICULATE RADIOACTIVITY

Background Airborne Radioactivity Concentrations

Poet et al. (1972) reported the average polonium-210 and lead-210 concentrations in ground level air to be 0.001 and 0.01

TABLE 8. WATER SAMPLES (SAMPLED 4/30/75)
RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

RADIONUCLIDE	WATER WELL #5 INCOMING (Total) pCi/l	COLD PIT WATER (LIQUID) pCi/l	COLD PIT WATER (SUSPENDED) pCi/g	WEST PLANT DISCHARGE WATER (Total) pCi/l
Ra - 226	0.70 ± 0.36	29 ± 2.0	2.8 ± 0.31	0.63 ± 0.31
Po - 210	< 0.82	ND*	5.2 ± 1.1	1.4 ± 0.93
U - 234	3.2 ± 0.4	39 ± 2.7	0.20 ± 0.031	9.6 ± 0.9
U - 235	0.12 ± 0.08	1.3 ± 0.2	< 0.0077	0.35 ± 0.10
U - 238	1.2 ± 0.2	39 ± 2.7	0.23 ± 0.038	6.2 ± 0.65
Th - 230	< 0.04	0.2 ± 0.1	5.1 ± 0.22	1.6 ± 0.30
Th - 232	< 0.04	< 0.033	0.031± 0.015	< 0.10
Pb - 210	1.2 ± 0.39	30 ± 1.6	0.45 ± 0.14	3.4 ± 1.2
Ra - 228	< 3.7	< 3.5	< 9.1	4.0 ± 3.5

*ND indicates non-detectable

TABLE 9. SCRUBBER DISCHARGES* (SAMPLED 4/30/75)
RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

RADIONUCLIDE	200 Plant Phos. Acid Scrubber (Suspended) pCi/g	200 Plant Phos. Acid Scrubber (Liquid) pCi/l	Calciner Scrubber (Suspended) pCi/g	Calciner Scrubber (Liquid) pCi/l	TSP East Scrubber (Suspended) pCi/g	TSP East Scrubber (Liquid) pCi/l	TSP West Scrubber (Suspended) pCi/g	TSP West Scrubber (Liquid) pCi/l
Ra - 226	2.5 ± 1.1	13 ± 1.4	33 ± 1.1	3.6 ± 0.72	10± 2.2	32 ± 2.1	22 ± 2.9	27 ± 1.9
Po - 210	--	45 ± 7.5	76 ± 14	0.67 ± 0.59	53± 8.6	ND	32 ± 7.1	ND
U - 234	--	37 ± 2.7	4.7 ± 1.6	6.8 ± 0.76	3.2± 0.50	43 ± 2.8	0.80 ± 0.20	56 ± 4.0
U - 235	--	1.1 ± 0.2	< 0.18	0.32 ± 0.12	< 0.10	1.6 ± 0.24	< 0.10	1.8 ± 0.24
U - 238	--	32 ± 2.2	5.0 ± 1.7	5.0 ± 0.60	3.5± 0.50	41 ± 2.8	1.0 ± 0.30	55 ± 3.6
Th - 230	--	0.70 ± 0.20	17 ± 0.60	0.20 ± 0.08	40± 2.2	16 ± 0.92	18 ± 1.4	32 ± 1.3
Th - 232	--	< 0.033	0.34 ± 0.080	< 0.04	0.40± 0.20	0.24 ± 0.12	0.40 ± 0.20	0.32 ± 0.12
Pb - 210	--	2.2 ± 0.42	3.1 ± 0.38	2.8 ± 0.59	6.7± 1.4	24 ± 1.4	3.1 ± 1.1	40 ± 1.7
Ra - 228	< 10	5.9 ± 3.6	1.0 ± 0.89	< 3.4	< 10	< 3.5	< 11	< 3.3

*ND indicates non-detectable; blanks indicate no data

pCi/m^3, respectively. NCRP (November, 1975) summarizes reports by geographic areas of the normal polonium-210 and lead-210 air concentrations. In Colorado, the reported polonium-210 concentrations ranged from 0.00005 to 0.003 pCi/m^3, whereas the lead-210 ranged from 0.001 to 0.021 pCi/m^3. The highest reported lead-210 concentrations were 0.026 pCi/m^3 for several locations in Illinois and Utah.

NCRP (November, 1975) also discusses the background level of airborne concentrations of the other naturally-occurring radionuclides. Natural uranium concentrations ranged from 120 aCi/m^3 (one attocurie is 10^{-18} curie or 10^{-6} pCi) near Chicago, Illinois, to a reported high of about 400 aCi/m^3 for several sites in New York State. Typical radium-226 concentrations are usually less than 100 aCi/m^3. Sedlet et al. (1973) reported thorium-232 and thorium-230 concentrations of 30 and 45 aCi/m^3, respectively, near Chicago, Illinois.

In-Plant Air Sampler

Air samples were obtained in the in-plant areas using a portable air sampling unit.* This unit has a carbon vane vacuum pump with a regulator which permits constant air flow sampling. Usually, sample collections were made at two cubic feet per minute (CFM) using a 47-millimeter diameter, Type E glass fiber filter. This corresponds to a linear flow rate of about 106 feet per minute. The air sample dust load was determined by measuring the mass of material collected on each filter.

Airborne particulate sampling was conducted in several working areas within the Wet Process Plant using the portable air sampler. The radioactivity concentrations of the naturally-occurring radionuclides are usually expressed as picocuries per

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* Regulated Air Sampler, Model RAS-1, Eberline Instrument Corporation, Santa Fe, New Mexico

cubic meter of sampled air (pCi/m^3). Dust loading of the air filter samples was also determined and the specific activity of the dust, expressed as pCi/g, is also given. The solubility of airborne particulate matter was not determined in this study.

Gross Versus Net Results

Eadie and Bernhardt (1976) have reported that radiochemical analyses of blank glass fiber filters (4-inch diameter) indicate appreciable quantities of naturally-occurring radioactivity. Such activity may be due to the composition of the filter media itself, or due to contaminants in reagents, glassware, or other analytical equipment. The analytical sensitivity of the radiochemical techniques may also mask the identification of the true source of such low levels of radioactivity.

Table 10 presents data on the radioactivity content of blank glass fiber filters (4-inch diameter) and the extrapolated activity content for the 47-mm diameter filters which were used in this study. In order to account for this radioactivity content associated with blank filter analyses, the appropriate blank filter activity (Table 10) has been subtracted from the measured gross analytical result to obtain a "net" result. No blank subtractions have been made for the three radionuclides (radium-228, polonium-210, and lead-210) which are at the analytical minimum detectable activity (MDA) levels. Gross analytical results for all of the in-plant air samples are shown in Appendix B.

TABLE 10. RADIOACTIVITY CONTENT OF BLANK GLASS FIBER FILTERS (pCi per filter)[+]

Radionuclide	4-inch Diameter[*]	47-mm Diameter[**]
Ra-226	0.35 ± 0.09	0.07 ± 0.019
Po-210	<0.17	<0.036
U-234	0.10 ± 0.03	0.021± 0.0064
U-235[***]	(0.0035 ± 0.0010)	(0.00075± 0.00021)
U-238	0.08 ± 0.02	0.017± 0.0043
Th-230	0.20 ± 0.08	0.043± 0.017
Th-232	0.13 ± 0.02	0.028± 0.0043
Pb-210	<0.32	<0.068
Ra-228	<1.6	<0.34

+ Average pCi per filter with standard error term about this mean based on the t-distribution at the 95 percent confidence level.

* Taken from Eadie and Bernhardt, 1976.

** Extrapolated value based on the area ratio between 47-mm and 4-inch diameter filters of 0.214 times the 4-inch diameter activity content. Average 47-mm filter mass ± two standard deviations was 0.1249 ± 0.0013 grams.

***Calculated U-235 content based on U-235 to U-238 natural activity ratio of 1:21.45 (0.0466).

In-Plant Air Sampler Net Results

Tables 11 to 16 present the net radionuclide concentrations obtained for the various in-plant sampling locations. As previously noted, the lead-210 results are underestimated by up to a factor of five, which results in a potential overestimate of about a factor of ten or more for the polonium-210 results. Data which narrows this uncertainty for specific samples are given in Appendix C.

Of all the in-plant samples, the highest airborne radioactivity was measured in the area of the No. 3 Calciner (Table 12). There, the measured airborne concentrations were orders of magnitude greater than results obtained in the Technical Building Library (Table 16). Although the library had the lowest airborne radioactivity measured in this study, even these levels were slightly elevated compared to reported background levels as discussed above.

Specific Activity of Airborne Particulate Matter

The specific activity of airborne particulate matter (reported in units of pCi/g) was also determined for the portable air sampler filters. The highest polonium-210 activity was 371 ± .46 pCi/g in the control room of the Calciner Building (Table 12) with roughly one-half of the sampling locations having essentially a non-detectable polonium-210 level (Tables 11, 13, 14, 15, and 16). The highest radium-226 activity of 41 ± 11 pCi/g was also obtained in the Calciner Building (Table 12) and the lowest activity was less than 2.1 pCi/g in the Technical Building Library (Table 16). These results for the Calciner Building also show possible disequilibrium between the individual radionuclides of the uranium-238 decay series, the most noticeable being the polonium-210 and thorium-230 contents compared to the other decay chain members. (Thorium-232 and radium-228 are members of a different decay series.) It should be recognized that the

TABLE 11. AIRBORNE PARTICULATE SAMPLING
NET RADIONUCLIDE CONCENTRATIONS ± TWO-SIGMA COUNTING ERROR TERM

RADIONUCLIDE	GRINDER MILL No. 7* 0906 to 1322 hrs. (4/30/75) pCi/m^3	pCi/g	ORE UNLOADING & STORAGE** 0945 to 1435 hrs. (5/1/75) pCi/m^3	pCi/g	GYPSUM PILE*** 0930 to 1442 hrs. (5/1/75) pCi/m^3
Ra - 226	0.23 ± 0.031	36 ± 4.9	0.16 ± 0.03	29 ± 5.6	0.10 ± 0.0046
Po - 210+	0.67 ± 0.27	100 ± 43	ND	ND	0.22 ± 0.062
U - 234	0.12 ± 0.017	19 ± 2.8	0.076 ± 0.014	14 ± 2.5	0.0072 ± 0.0012
U - 235	< 0.0028	< 0.44	0.0084 ± 0.0045	1.5 ± 0.81	< 0.00016
U - 238	0.13 ± 0.018	21 ± 2.9	0.092 ± 0.015	17 ± 2.8	0.0079 ± 0.0011
Th - 230	0.29 ± 0.034	46 ± 5.4	0.17 ± 0.027	32 ± 5.0	0.032 ± 0.0028
Th - 232	< 0.0059	< 0.93	0.0080 ± 0.0064	1.4 ± 1.3	0.00057 ± 0.00046
Pb - 210+	0.19 ± 0.068	31 ± 11	0.24 ± 0.071	45 ± 13	0.076 ± 0.006
Ra - 288+	< 0.10	< 16	< 0.10	< 19	< 0.0055

*Sampled Volume of 16.6m^3 with a dust particulate load of 0.1044g.

**Sampled Volume of 16.6m^3 with a dust particulate load of 0.0902g.

***Sampled Volume of 333m^3. Collected on 8 x 10-inch glass fiber filter.

+Gross analytical result.

TABLE 12. AIRBORNE PARTICULATE SAMPLING
NET RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

RADIONUCLIDE	CALCINER - CONTROL ROOM* 1058 to 1520 hrs. (4/28/75) pCi/m^3	pCi/g	#3 CALCINER** 1154 to 1335 hrs. (4/28/75) pCi/m^3	pCi/g
Ra - 266	0.069 ± 0.019	41 ± 11	1.6 ± 0.14	32 ± 2.8
Po - 210+	0.60 ± 0.26	371 ± 146	9.8 ± 1.2	195 ± 24
U - 234	0.029 ± 0.0086	16 ± 5.0	1.0 ± 0.10	20 ± 2.0
U - 235	< 0.0025	< 1.5	0.092 ± 0.025	1.8 ± 0.50
U - 238	0.034 ± 0.0096	19 ± 5.6	1.0 ± 0.10	20 ± 2.0
Th - 230	0.06 ± 0.017	35 ± 9.5	1.8 ± 0.16	37 ± 3.1
Th - 232	< 0.002	< 1.2	0.071 ± 0.038	1.4 ± 0.74
Pb - 210+	0.077± 0.065	42 ± 36	< 0.14	< 2.8
Ra - 228+	< 0.10	< 61	< 0.31	< 6.2

*Sampled Volume of 16.5m^3, with a dust particulate load of 0.0285g.

**Sampled Volume of 5.44m^3, with a dust particulate load of 0.2742g.

+ Gross analytical result

TABLE 13. AIRBORNE PARTICULATE SAMPLING in the 300 PHOSPHORIC ACID PLANT++

NET RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

	ABOVE DIGESTER*		OUTSIDE CONTROL ROOM**		CONTINUOUS FILTER*** (LANDSKRONA)		CONTROL ROOM****	
	1016 to 1157 hrs. (4/28/75)		1129 to 1452 hrs. (4/28/75)		1020 to 1233 hrs. (4/28/75)		1227 to 1537 hrs. (4/28/75)	
RADIONUCLIDE	pCi/m^3	pCi/g	pCi/m^3	pCi/g	pCi/m^3	pCi/g	pCi/m^3	pCi/g
Ra - 226	0.39 ± 0.12	25 ± 7.7	0.078 ± 0.029	12 ± 4.3	< 0.016	< 5.6	0.026 ± 0.018	16 ± 11
Po - 210+	ND	ND	ND	ND	< 0.12	< 48	< 0.43	< 276
U - 234	0.24 ± 0.059	16 ± 3.7	0.12 ± 0.022	18 ± 3.3	0.06 ± 0.018	22 ± 6.8	0.016 ± 0.0093	10± 6.0
U - 235	< 0.023	< 1.5	0.013 ± 0.0078	2.0 ± 1.2	< 0.0034	< 1.3	< 0.0044	< 2.9
U - 238	0.18 ± 0.055	12 ± 3.5	0.14 ± 0.025	21 ± 3.7	< 0.0022	< 0.81	0.016 ± 0.0091	11± 6.0
Th - 230	0.43 ± 0.10	28 ± 6.5	0.22 ± 0.038	33 ± 5.9	0.069 ± 0.026	25 ± 9.1	0.022 ± 0.014	15± 9.0
Th - 232	0.039 ± 0.034	2.5 ± 2.3	< 0.0067	< 1.1	< 0.0063	< 2.3	< 0.0014	< 0.91
Pb - 210+	0.82 ± 0.40	54 ± 26	0.36 ± 0.13	55 ± 19	0.18 ± 0.14	66 ± 49	0.16 ± 0.12	100± 78
Ra - 228+	< 0.49	< 32	< 0.17	< 26	< 0.19	< 68	< 0.17	< 110

+Gross analytical results

++ND indicates non-detectable.

*Sampled volume of $3.02m^3$, with a dust particulate load of 0.0462g.

**Sampled volume of $10.1m^3$, with a dust particulate load of 0.0661g.

***Sampled volume of $7.91m^3$, with a dust particulate load of 0.0215g.

****Sampled volume of $8.93m^3$, with a dust particulate load of 0.0134g.

TABLE 14. AIRBORNE PARTICULATE SAMPLING
NET RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

RADIONUCLIDE	TSP STORAGE AREA* 1020 to 1412 hrs. (4/29/75) pCi/m^3	pCi/g	TSP DRYER** 0831 to 1200 hrs. (4/30/75) pCi/m^3	pCi/g	ACIDULATION TSP DISCHARGE*** 0811 to 1207 hrs. (4/30/75) pCi/m^3	pCi/g
Ra - 226	0.014 ± 0.0086	10 ± 4.9	0.56 ± 0.057	17 ± 1.7	0.065 ± 0.022	24 ± 7.8
Po - 210+	< 0.17	< 144	1.4 ± 0.51	39 ± 15	ND	ND
U - 234	0.013 ± 0.0058	8.9 ± 4.1	2.1 ± 0.17	63 ± 5.1	0.066 ± 0.015	26 ± 5.9
U - 235	< 0.0027	< 2.0	0.054 ± 0.014	1.6 ± 0.43	0.0039 ± 0.0038	1.6 ± 1.5
U - 238	0.013 ± 0.0057	9.1 ± 4.1	1.4 ± 0.12	41 ± 3.6	0.048 ± 0.012	18 ± 4.8
Th - 230	0.011 ± 0.0094	7.8 ± 6.1	2.6 ± 0.10	78 ± 3.7	0.15 ± 0.029	55 ± 11
Th - 232	< 0.00086	< 0.58	0.023 ± 0.012	0.66 ± 0.35	0.0088 ± 0.0086	3.3 ± 3.2
Pb - 210+	0.12 ± 0.079	85 ± 57	0.68 ± 0.12	20 ± 3.6	0.14 ± 0.094	53 ± 35
Ra - 228+	< 0.10	< 84	< 0.13	< 3.8	0.31 ± 0.15	110 ± 55

*Sampled volume of 14.4m^3, with a dust particulate load of 0.0197g.

**Sampled volume of 11.8m^3, with a dust particulate load of 0.3998g.

***Sampled volume of 12.6m^3, with a dust particulate load of 0.0332g.

+Gross analytical result. ND indicates non-detectable.

TABLE 15. AIRBORNE PARTICULATE SAMPLING
NET RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

	200 AMMOPHOS PLANT - DRYER* (ground floor) 0935 to 1201 hrs. (4/29/75)		100 AMMOPHOS PLANT - DRYER** (ground floor) 0943 to 1200 hrs. (4/29/75)	
RADIONUCLIDE	pCi/m^3	pCi/g	pCi/m^3	pCi/g
Ra - 226	0.077 ± 0.03	4.1±1.6	0.089 ± 0.033	10 ± 3.7
Po - 210+	ND	ND	< 0.18	< 20
U - 234	0.37 ± 0.048	20±2.6	0.25 ± 0.036	29 ± 4.1
U - 235	0.0077 ±0.0068	0.42±0.37	0.0081 ± 0.0065	0.93 ± 0.75
U - 238	0.35 ±0.046	18±2.4	0.27 ± 0.038	31 ± 4.4
Th - 230	0.57 ±0.066	31±3.3	0.54 ± 0.072	62 ± 8.2
Th - 232	<0.0053	<0.29	< 0.0075	< 0.89
Pb - 210+	0.34 ±0.14	18±7.6	0.16 ± 0.14	18 ± 16
Ra - 228+	<0.21	<11	< 0.22	< 25

*Sampled Volume of $8.2m^3$, with a dust particulate load of 0.1525g.

**Sampled Volume of $7.9m^3$, with a dust particulate load of 0.0686g.

+Gross analytical results. ND indicates non-detectable.

TABLE 16. AIRBORNE PARTICULATE SAMPLING
NET RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

RADIONUCLIDE	TECH. BLDG. LIBRARY (STORAGE)* 1036 to 1600 hrs. (4/29-30/75) pCi/m^3	pCi/g	200 AMMOPHOS PLANT** 18-46-0 Storage 1231 to 1600 hrs. (4/29/75) pCi/m^3	pCi/g	100 AMMOPHOS PLANT*** 11-54-0 Storage 1248 to 1607 hrs. (4/29/75) pCi/m^3	pCi/g
Ra - 226	<0.00043	< 2.1	0.021 ± 0.015	2.5 ± 1.6	0.021 ± 0.014	5.3 ± 3.3
Po - 210+	ND	ND	ND	ND	ND	ND
U - 234	0.0025 ± 0.0013	11 ± 5.2	0.24 ± 0.032	28 ± 3.8	0.15 ± 0.023	36 ± 5.5
U - 235	< 0.00055	< 2.5	0.0087 ± 0.0056	0.99 ± 0.67	0.0042 ± 0.004	0.99 ± 0.95
U - 238	0.0017 ± 0.0012	7.6 ± 5.0	0.24 ± 0.032	28 ± 3.7	0.16 ± 0.024	37 ± 5.6
Th - 230	0.0036 ± 0.0025	19 ± 9.1	0.48 ± 0.052	54 ± 6.1	0.28 ± 0.039	67 ± 9.4
Th - 232	< 0.000073	< 0.34	< 0.0057	< 0.70	0.015 ± 0.01	3.6 2.5
Pb - 210+	0.048 ± 0.014	240 ± 71	0.17 ± 0.091	20 ± 11	0.14 ± 0.094	34 ± 22
Ra - 228+	< 0.019	< 95	< 0.14	< 17	< 0.12	< 30

*Sampled volume of 19.2m^3, with a dust particulate load of 0.0179g.

**Sampled volume of 12.1m^3, with a dust particulate load of 0.1042g.

***Sampled volume of 12.3m^3, with a dust particulate load of 0.0507g.

+Gross analytical result. ND indicates non-detectable.

apparent disequilibrium can be partially accounted for by the errors in the polonium and lead-210 data and analytical uncertainties (see Appendix C). The results for #3 calciner do indicate the strong liklihood of excess polonium-210 (see Appendix C).

Specific activity determinations of the airborne particulate matter may also be compared to the results of analyses of the raw material input to the wet process (Table 3). The highest polonium-210 content of the input ore was 48 ± 5.9 pCi/g (Conda Mine-low grade ore). The specific activity determinations of the airborne particulate samples from the Calciner Building indicate potentially higher polonium-210 than the input ore concentration. The specific activity concentrations for the other radionuclides in the input products are generally comparable to the airborne particulate sample results for all locations sampled.

PARTICLE SIZE CHARACTERIZATION

Cascade Impactor Sampling

High volume cascade impactors* were used to measure the size distribution of airborne particulate matter for both the indoor and outdoor environments. The impactor filter stages attach to a high volume air sampler** which is electronically controlled*** to operate at a flow rate of 40 cubic feet per minute (CFM). This corresponds to a linear air velocity of about 72 feet per minute through the final 8 x 10-inch filter stage.

Whatman #41 paper filters were used for each impaction stage. Glass fiber (Type E) was used for the final 8 x 10-inch filter. Typical particle size ranges for each filter stage, as reported by the manufacturer, are:

* Model 252 Series, six stage, cascade impactor from Tech Ecology, Inc. (Now produced and marketed by Sierra Instruments, Inc.)

** Model GMWL-2000, High Volume Air Sampling System from General Metal Works, Inc.

***Model 310/310A, High Volume Constant Flow Controller from Sierra Instruments, Inc.

Stage No.	Equivalent Aerodynamic Diameter at 50 Percent Collection Efficiency (micrometer)
1	Greater than 7.2 μm
2	3.0 - 7.2
3	1.5 - 3.0
4	0.95- 1.5
5	0.49- 0.95
Final Filter	Less than 0.49

"Equivalent aerodynamic diameter" is defined as the size of a spherical particle of unit density which has the same terminal settling velocity as the sampled particle. Radioactivity analysis of each impactor stage provides an indication of the activity content for the various particle size ranges.

Gross Results of Impactor Sampling

The cascade impactor sampler was used at four locations in the Wet Process Plant - the Calciner, the ore unloading and storage area, the 200 Ammophos Plant (18-46-0 storage area), and at the gypsum pile. Gross analytical results for these samples are presented in Tables 17 to 20. Tables 21 and 22 show the results of radiochemical analyses of blank impactor filters - the slotted Whatman #41 paper and the 8 x 10-inch glass fiber filters, respectively. Since most of the blank slotted paper filters contained radioactivity concentrations at the minimum detectable activity (MDA) levels for the analysis, a blank filter subtraction has not been performed for all the radionuclides for the impactor samples. As noted in subsequent discussions, tabulations of net results have been used in some of the data plots. The previously noted reservations concerning the lead-210 and polonium-210 data also apply to these results (i.e., lead-210 may be low by a factor of five or more and polonium-210 high by a factor of ten). Data for narrowing the uncertainty on specific samples are given in Appendix C.

TABLE 17. CASCADE IMPACTOR SAMPLING IN THE ORE UNLOADING & STORAGE AREA*
GROSS RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM, IN pCi/m^3

RADIONUCLIDE	FILTER #1	FILTER #2	FILTER #3	FILTER #4	FILTER #5	FINAL FILTER	ACTIVITY SUMMATION FOR ALL FILTERS
Ra - 226	0.0073 ± 0.0051	0.014 ± 0.006	0.019 ± 0.0067	0.027 ± 0.0082	0.055 ± 0.011	0.22 ± 0.022	0.34
Po - 210	0.16 ± 0.14	ND	ND	ND	ND	0.39 ± 0.17	0.55
U - 234	0.04 ± 0.01	0.019 ± 0.0056	0.025 ± 0.0076	0.013 ± 0.0039	0.03 ± 0.01	0.15 ± 0.015	0.28
U - 235	< 0.0025	< 0.0018	< 0.0025	< 0.0014	< 0.0025	0.0064 ± 0.0027	< 0.017
U - 238	0.04 ± 0.01	0.022 ± 0.006	0.018 ± 0.0076	0.014 ± 0.0042	0.023 ± 0.0076	0.15 ± 0.016	0.27
Th - 230	0.053 ± 0.013	0.04 ± 0.01	0.028 ± 0.01	0.032 ± 0.0091	0.028 ± 0.013	0.34 ± 0.028	0.52
Th - 232	< 0.0025	< 0.0033	< 0.0025	0.005 ± 0.0049	< 0.0025	0.011 ± 0.0052	< 0.027
Pb - 210	0.07 ± 0.035	0.074 ± 0.035	0.088 ± 0.036	0.081 ± 0.036	0.083 ± 0.036	0.21 ± 0.043	0.61
Ra - 228	< 0.054	< 0.054	< 0.052	< 0.057	< 0.052	< 0.053	< 0.32
Percent of Filter Stage to Total Activity	15%	6.5%	8.0%	7.9%	9.4%	52%	

*Sampled volume of 31.7m^3 from 1347 to 1415 hrs. (5/1/75). ND indicates non-detectable.

TABLE 18. CASCADE IMPACTOR SAMPLING IN THE CALCINER BUILDING*
GROSS RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM, IN pCi/m^3

RADIONUCLIDE	FILTER #1	FILTER #2	FILTER #3	FILTER #4	FILTER #5	FINAL FILTER	ACTIVITY SUMMATION FOR ALL FILTERS
Ra - 226	0.031 ± 0.0081	0.039 ± 0.0094	0.0070 ± 0.0049	<0.0043	0.013 ± 0.0055	0.15 ± 0.017	0.24
Po - 210	0.27 ± 0.15	0.25 ± 0.083	0.69 ± 0.39	0.24 ± 0.13	0.26 ± 0.16	0.75 ± 0.16	2.5
U - 234	0.071 ± 0.012	0.017 ± 0.0044	0.029 ± 0.0071	0.048 ± 0.0071	0.043 ± 0.0095	0.10 ± 0.012	0.31
U - 235	<0.0024	<0.0012	<0.0024	0.0048 ± 0.0024	<0.0024	0.003 ± 0.002	<0.016
U - 238	0.086 ± 0.012	0.021 ± 0.0048	0.033 ± 0.0071	0.036 ± 0.0071	0.040 ± 0.0095	0.10 ± 0.012	0.32
Th - 230	0.10 ± 0.019	0.044 ± 0.01	0.062 ± 0.012	0.045 ± 0.0095	0.045 ± 0.012	0.20 ± 0.02	0.50
Th - 232	< 0.0024	0.0040 ± 0.0038	0.0048 ± 0.0024	0.0048 ± 0.0024	<0.0024	0.0056 ± 0.0038	<0.024
Pb - 210	0.088 ± 0.033	0.023 ± 0.019	<0.03	<0.032	0.050 ± 0.032	0.12 ± 0.036	<0.34
Ra - 228	<0.051	<0.051	<0.056	<0.058	<0.065	<0.056	<0.34
Percent of Filter Stage to Total Activity	15%	9.8%	20%	10%	11%	32%	

*Sampled volume of 33.6m^3 from 1341 to 1411 hrs. (4/28/75) near the #3 Calciner, next to the hot cyclone-2 levels above the control room.

TABLE 19. CASCADE IMPACTOR SAMPLING IN THE 200 AMMOPHOS PLANT (18-46-0 storage area)*
GROSS RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM, pCi/m^3

RADIONUCLIDE	FILTER #1	FILTER #2	FILTER#3	FILTER #4	FILTER #5	FINAL FILTER	ACTIVITY SUMMATION FOR ALL FILTERS
Ra - 226	0.0062 ± 0.0044	0.0057 ± 0.0045	0.0093 ± 0.0046	0.0088 ± 0.0047	0.007 ± 0.0049	0.027 ± 0.0078	0.064
Po - 210	<0.064	<0.12	ND	ND	ND	0.12 ± 0.11	<0.30
U - 234	0.42 ± 0.036	0.34 ± 0.029	0.089 ± 0.012	0.053 ± 0.0097	0.034 ± 0.017	0.10 ± 0.012	1.0
U - 235	0.012 ± 0.0072	0.0097 ± 0.0048	0.0048 ± 0.0024	<0.0024	<0.0024	0.0036 ± 0.002	<0.035
U - 238	0.43 ± 0.036	0.35 ± 0.029	0.087 ± 0.012	0.06 ± 0.0097	0.029 ± 0.0072	0.092 ± 0.011	1.0
Th - 230	0.83 ± 0.046	0.52 ± 0.034	0.13 ± 0.017	0.094 ± 0.017	0.06 ± 0.012	0.20 ± 0.021	1.8
Th - 232	0.0097 ± 0.0048	<0.0048	<0.0024	<0.0024	<0.0024	<0.0023	<0.024
Pb - 210	0.11 ± 0.037	0.073 ± 0.035	0.10 ± 0.037	0.07 ± 0.036	0.097 ± 0.037	<0.032	<0.48
Ra - 228	<0.054	<0.055	<0.054	<0.056	<0.053	<0.055	<0.33
Percent of Filter Stage to Total Activity	39%	30%	9.5%	6.9%	5.7%	13%	

*Sampled volume of 33.1m^3 from 1429 to 1459 hrs. (4/29/75). ND indicates non-detectable.

TABLE 20. CASCADE IMPACTOR SAMPLING AT THE GYPSUM PILE*
GROSS RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM, in pCi/m^3

RADIONUCLIDE	FILTER #1	FILTER #2	FILTER #3	FILTER #4	FILTER #5	FINAL FILTER	ACTIVITY SUMMATION FOR ALL FILTERS
Ra - 226	0.0043 ± 0.0012	0.0045 ± 0.0012	0.0053 ± 0.0013	0.0078 ± 0.0016	0.0063 ± 0.0014	0.15 ± 0.0067	0.18
Po - 210	ND	ND	ND	ND	ND	0.20 ± 0.074	0.20
U - 234	0.00074 ± 0.00037	0.00048 ± 0.00031	0.0011 ± 0.00053	0.0011 ± 0.00048	0.00055 ± 0.00034	0.012 ± 0.0015	0.016
U - 235	<0.00014	<0.0001	<0.00014	<0.00012	<0.00019	0.00031 ± 0.00023	< 0.0010
U - 238	0.00086 ± 0.0004	0.00074 ± 0.00039	0.0007 ± 0.00036	<0.00012	0.00079 ± 0.00044	0.014 ± 0.0016	< 0.017
Th - 230	0.0026 ± 0.001	0.0036 ± 0.0012	0.0024 ± 0.00088	0.0023 ± 0.00086	0.003 ± 0.00096	0.036 ± 0.0033	0.050
Th - 232	<0.00024	<0.00039	<0.00018	<0.00034	<0.00039	0.0011 ± 0.00062	< 0.0026
Pb - 210	0.011 ± 0.0051	0.011 ± 0.0052	0.017 ± 0.0055	0.015 ± 0.005	0.017 ± 0.0051	0.11 ± 0.016	0.18
Ra - 228	<0.0072	<0.0073	<0.0073	<0.0072	<0.0079	<0.0078	< 0.045
Percent of Filter Stage to Total Activity	3.9%	4.1%	4.9%	4.9%	5.2%	77%	

*Sampled volume of 231m^3 from 0943 to 1312 hrs. (5/1/75). ND indicates non-detectable.

TABLE 21. RADIOACTIVITY CONTENT OF BLANK IMPACTOR WHATMAN #41 PAPER FILTER (SLOTTED)
(pCi ± TWO-SIGMA COUNTING ERROR TERM PER FILTER)

Filter	Ra-226	Po-210	U-234	U-235*	U-238	Th-230	Th-232	Pb-210	Ra-228
1	<0.13	<5.1	<0.36	<0.014	<0.31	-	-	<1.3	<1.7
2		<3.1	<0.95	<0.023	<0.50	<0.068	<0.068	<1.2	<2.0
3	0.23 ± 0.17	12 ± 6.4	1.2 ±0.85	<0.023	<0.50	<0.070	<0.034	<1.2	<2.2
4	0.37 ± 0.17	7.8 ± 4.2	<0.67	<0.030	<0.64	<0.045	<0.034	<1.1	<2.0
5	0.17 ± 0.14	10 ± 5.0	0.70 ±0.62	<0.033	<0.70	<0.062	<0.048	<1.1	<2.0
Average of All Results**	<0.23	<7.6	<0.78	<0.025	<0.53	<0.061	<0.046	<1.2	<2.0

*U-235 calculated based on natural U-235 to U-238 activity ratio of 1 to 21.45 (0.0466). Blanks indicate no data.
**All average results are reported as less than values since most of the analytical results of the individual filter are less than values.

TABLE 22. RADIOACTIVITY CONTENT OF BLANK GLASS FIBER FILTERS (8 BY 10-INCH)
(pCi ± TWO-SIGMA COUNTING ERROR TERM PER FILTER)

Filter	Ra-226	Po-210	U-234	U-235*	U-238	Th-230	Th-232
1	0.21 ± 0.08	0.64 ± 0.38	0.29 ± 0.10	0.012 ± 0.0045	0.26 ± 0.096	0.33 ± 0.11	0.14 ± 0.072
2	0.15 ± 0.07	<0.53	0.34 ± 0.12	0.014 ± 0.0051	0.31 ± 0.11	0.33 ± 0.13	0.15 ± 0.08
3	0.57 ± 0.13	LOST	0.35 ± 0.16	0.018 ± 0.0084	0.38 ± 0.18	0.44 ± 0.18	0.16 ± 0.15
4	0.32 ± 0.10	<0.54	0.36 ± 0.13	0.017 ± 0.0061	0.36 ± 0.13	0.40 ± 0.14	0.18 ± 0.088
5	0.17 ± 0.07	<0.58	0.25 ± 0.11	0.0079 ± 0.0041	0.17 ± 0.088	0.32 ± 0.13	0.18 ± 0.096
6	0.23 ± 0.08	<0.58	0.30 ± 0.13	0.0075 ± 0.0061	0.16 ± 0.13	0.22 ± 0.11	0.13 ± 0.072
Average of All Results**	0.28 ± 0.16	<0.57	0.32 ± 0.04	0.013 ± 0.0044	0.27 ± 0.094	0.34 ± 0.076	0.16 ± 0.021

*U-235 calculated based on natural U-235 to U-238 activity ratio of 1 to 21.45 (0.0466).
**Average of all results with standard error about this mean based in the t-distribution at the 95 percent confidence level.

Table 23 provides a summary of the gross results of the cascade impactor sampling for the four sampling locations. Except for the results from the 200 Ammophos Plant the majority of radioactivity (ranging from 32 to 77 percent of the arithmetic total) was measured in the particle size fraction less than 0.49 μm. The average of all four sampling locations indicates that roughly 52 percent of the arithmetic total airborne radioactivity was in the sub-micron particle size range.

Comparison of Air Sampling Results

The activity summation for all filters of the cascade impactor may be compared to the activity concentrations determined from the portable air sampler for the same sampling location. Good agreement has been obtained between the results of the radioactivity determinations using these two different sampling systems, even though the samples were not necessarily obtained during simultaneous time periods.

For the Ore Unloading and Storage Area, impactor results (Table 17) are roughly two to three times the activity concentrations as measured using the portable air sampler (third column of Table 11). In both cases, decay chain members appear to be in equilibrium.

In the Calciner Building, the cascade impactor results (Table 18) are about one-third of the activity measured using the portable air sampler (third column of Table 12), except for the radium-226 which was one-seventh of the portable air sampler result. Of the four locations sampled for airborne radioactivity using both sampling systems, only the Calciner Building impactor results (Table 18) showed less airborne radioactivity than did the results obtained using the portable air sampler (Table 12). Of all the sampling locations, the highest airborne radioactivity measurements were obtained in the Calciner Building, regardless of the air sampling system used. (This conclusion recognizes the previously denoted uncertainties for the lead-210 and polonium-210 results.)

TABLE 23. SUMMARY OF GROSS RESULTS OF THE CASCADE IMPACTOR SAMPLING — PERCENTAGE OF TOTAL RADIOACTIVITY PER PARTICLE SIZE RANGE

Location	Greater than 7.2 μm	3.0 to 7.2 μm	1.5 to 3.0 μm	0.95 to 1.5 μm	0.49 to 0.95 μm	Less than 0.49 μm
ORE LOADING AND STORAGE AREA	15	9.8	20	10	11	32
CALCINER BUILDING	15	6.5	8.0	7.9	9.4	52
200 AMMOPHOS PLANT (18-46-0 storage area)	39	30	9.5	6.9	5.7	13
GYPSUM PILE	3.9	4.1	4.9	4.9	5.2	77
ARITHMETIC AVERAGE PERCENTAGE	18	13	11	7.4	7.8	44

Impactor sampling in the 18-46-0 storage area of the 200 Ammophos Plant (Table 19) showed about three times the airborne radioactivity as obtained using the portable air sampler (third column of Table 16). Excellent agreement was obtained for the results from both sampling systems at the Gypsum Pile (Tables 20 and the fifth column of Table 11).

Particle Size Distribution

Figures 2, 3, 4 and 5 present the log-normal probability plots of data from the cascade impactor sampling. The plotted points are based on both the gross activity and in some cases, the net activity (i.e., gross minus the appropriate activity associated with a blank filter). Only total uranium, thorium-230, and radium-226 data have been plotted. Data for the other remaining radionuclides were not plotted because of inadequate blank filter results or because the gross results were so low that meaningful net results could not be calculated. The actual points were calculated by allocating all the activity in all the stages subsequent to and including that particular stage to the specified size cut off for that stage (Elder et al., 1974). The manufacturer specified size cutoffs for the impactor at 40 cfm are 7.2, 3.0, 1.5, 0.95, and 0.49 micrometers (equivalent aerodynamic diameter). These are partially based on a study by Willeke (1975). Thus, in the actual calculations, the percent of the total activity on the 3.0, 1.5, 0.95, and 0.49 and final filter stages was designated as less than 7.2 micrometers. The percent of the total activity on the final filter was ascribed to the 0.49 micrometer stage, etc. The lines are based on hand-drawn estimates and not based on regression analysis.

The plotted distributions (Figures 2, 3, 4 and 5) have not been statistically tested to demonstrate the appropriateness of describing the data with a log-normal distribution. The log-probability plots are used as a tool to summarize the data and describe its central tendency.

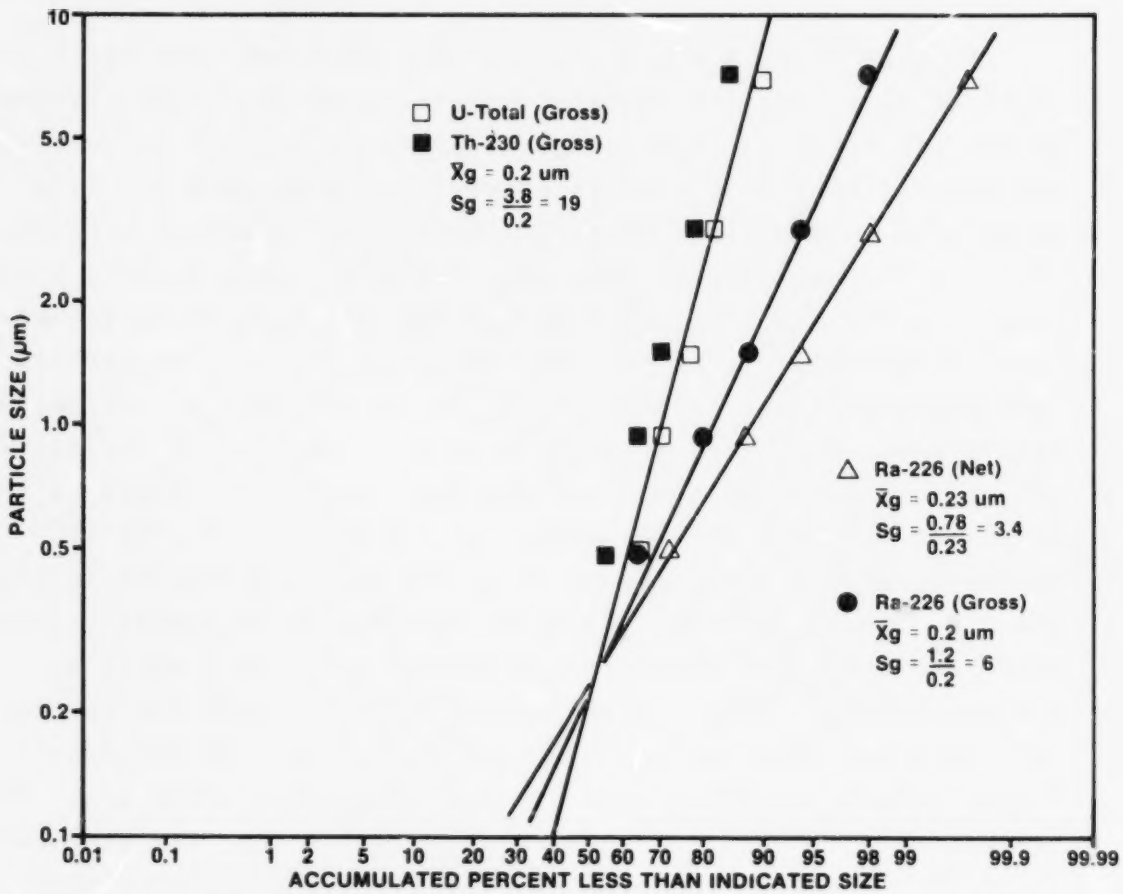

Figure 2. Ore unloading and storage area log-probability plot of the particle size distribution (Table 17).

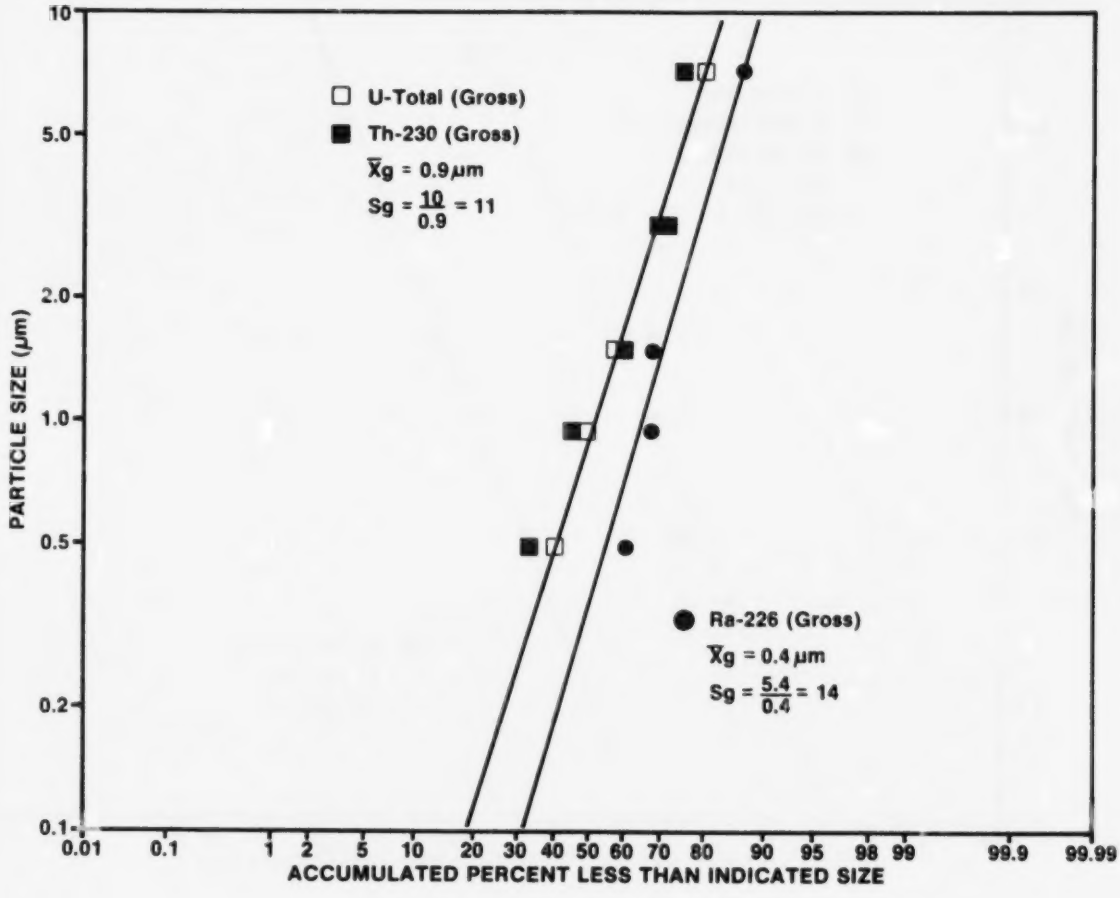

Figure 3. Calciner building log-probability plot of the particle size distribution (Table 18).

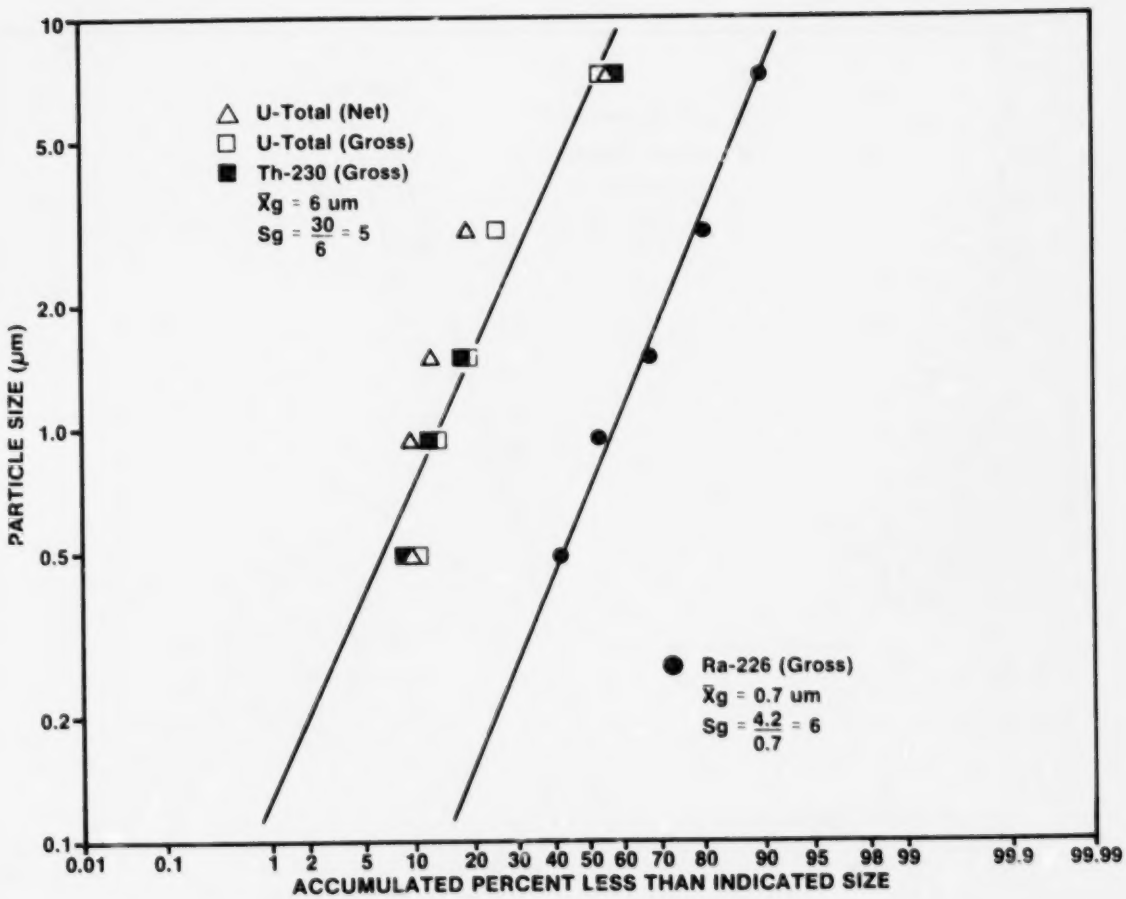

Figure 4. 200 Ammophos plant log-probability plot of the particle size distribution (Table 19).

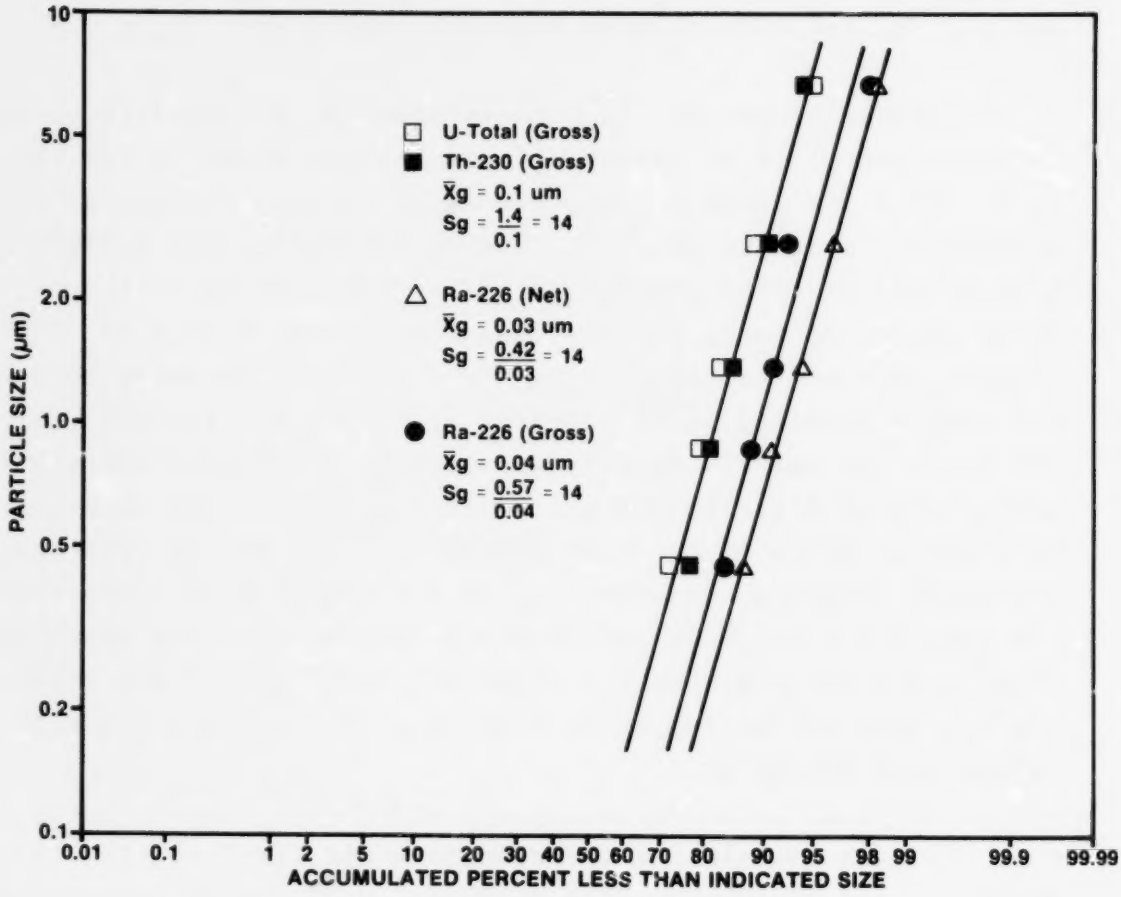

Figure 5. Gypsum pile log-probability plot of particle size distribution (Table 20).

The geometric mean ($\overline{X}_g$) is given by the fiftieth cumulative percent intercept. The geometric standard deviation (S_g) is indicated by the slope of the line and is usually calculated by dividing the particle diameter at the 84 percentile by the diameter at the 50 percentile. Due to the limited data and the uncertainty of analytical results, there is considerable uncertainty in the log-normal probability values. Many of the values have been rounded to one significant digit, and those that have not are not accurate to more than one significant digit.

In order to present as much inforamtion as possible, size distribution plots of the gross activity are given in Figures 2 to 5. When the measured sample activity was sufficient to allow calculation of reasonably accurate quantities of net activity for most of the impactor stages, the size distribution of the net activity results were also plotted (Figures 2, 4, and 5). The significance of the quantity of blank activity compared to the net sample activity is illustrated in Figure 2. Subtraction of the blank radium-226 activity resulted in a slightly different particle size distribution plot with essentially the same geometric mean value ($\overline{X}_g$ of 0.20 versus 0.23 μm) but an apparently different standard deviation (S_g of 6.0 versus 3.4). The thorium-230 sample activity was sufficiently greater than the appropriate blank activity such that the resulting "net" values are essentially identical to the gross distribution therefore the net values were not shown.

The data for the Ore Unloading and Storage Area, the Calciner Building, and the Gypsum Pile (Figures 2, 3, and 5, respectively) indicate geometric means of less than one micrometer. These results are within expected values for atmospheric and industrial dusts (Lee, 1972; Elder et al., 1974; and Willeke, 1975). Relatively large geometric means, indicating rather coarse particulate matter, was obtained in the 200 Ammophos Plant (Figure 4).

Geometric standard deviations (S_g) ranged from 3.4 to 19. These values are similar to, but slightly higher than reported values for ambient data (generally less than S_g of 10; Lee, 1972). These relatively high geometric standard deviations probably reflect the composite of several size distributions. That is, there may be several particle size distributions from the plant in conjunction with ambient dust. The results for the Ore Unloading and Storage Area (Figure 2) and the Calciner Building (Figure 3) show concentrations above ambient levels.

Knuth (1976), in his evaluation of the impactor, indicated that it tended to underestimate the size distribution and overestimate the geometric standard deviation. Thus, it is possible that this is also reflected in the data. Although Knuth's results indicated that this appeared to be more of a problem for the larger size distributions ($\bar{X}_g$ of 7 micrometers) but was less of a problem for distributions around 4 micrometers.

Results from Lee (1972) indicate average geometric standard deviations of up to 20 for ambient air. Elder et al., (1974) indicate geometric standard deviations in the thirties for processes using plutonium; thus, the values noted in this study appear to be realistic.

As shown in Figures 2 to 5, two different particle size distributions are implied by the data for total-uranium and thorium-230 data versus the radium-226 data. The geometric mean of the uranium and thorium data for the Calciner and the Gypsum Pile (Figures 3 and 5) are about twice that of the radium data and about a factor of 10 higher for the 200 Ammophos Plant (Figure 4). The geometric means are similar for the Ore Area (Figure 2), but the thorium and uranium data have a higher geometric standard deviation than the radium data.

STACK SAMPLING

Stack sampling was conducted in late September 1975 using the RAC Train Stacksamplr* and methods specified in the Federal Register, Volume 36, No. 247 (December 23, 1971). Representative samples were obtained from each process-type discharge stack; however, every plant discharge stack was not sampled.

Particulate matter was collected on a glass fiber filter (2.5-inch diameter) which was subsequently analyzed for natural radioactivity content. To determine the blank filter natural radioactivity content, two sets of unused filters were analyzed and these results are shown in Table 24. Also shown is the extrapolated filter content based on the area ratio between the 2.5-inch and the 4-inch diameter filters of 0.391 times the 4-inch diameter activity content as reported by Eadie and Bernhardt, (1976). These results show fairly good agreement between the measured and the extrapolated activity contents. The extrapolated filter content (last column of Table 24) has been used as a blank filter activity content which has been subtracted from the gross analytical result to obtain the reported "net result". No blank subtractions have been made for the three radionuclides (radium-228, polonium-210, and lead-210) which are at the analytical minimum detectable activity (MDA) level. The net results for the stack effluent discharge radioactivity concentrations are reported in Table 25.

The 100 Calciner Scrubber and the 100 Ammophos Reactor stacks were sampled, but the sampling was not done under isokinetic conditions. Thus, these results have not been reported. Assuming the results were not grossly in error (sampling was 25 and 35 percent outside of isokinetic), the concentrations of activity in these stacks (uranium-238 chain) were around 1 pCi/m^3. The flow rates for these stacks were similar to those given for the data in Table 25, indicating that a large proportion of the

* Research Appliance Corporation (RAC), Gibsonia, PA.

effluent was not missed. That is, more radioactivity is accounted for by the Mill Stacks and TSP Dryer than by the Calciner Scrubber and Ammophos Reactor.

TABLE 24. RADIOACTIVE CONTENT OF BLANK GLASS FIBER FILTER (2.5-INCH DIAMETER)*
(pCi/filter ± two-sigma counting error term)

Radionuclide	5-Filter Composite	6-Filter Composite	Average Blank Filter Content	Extrapolated Filter Content++
Ra-226	0.52 ± 0.14	0.42 ± 0.16	0.47 ± 0.21	0.14 ± 0.035
Po-210	0.29 ± 0.11	0.058 ± 0.015	0.17 ± 0.11	<0.066
U-234	0.060 ± 0.038	0.022 ± 0.018	<0.041	0.039 ± 0.012
U-235**	<0.0012	0.0012± 0.00079	<0.0012	0.0014± 0.00039
U-238	<0.026	0.025 ± 0.017	<0.026	0.031 ± 0.0078
Th-230	<0.075	0.089 ± 0.040	<0.082	0.078 ± 0.031
Th-232	<0.080	0.029 ± 0.023	<0.055	0.051 ± 0.0078
Pb-210	No data	<0.088	<0.088	<0.13
Ra-228	<0.81	No data	<0.81	<0.62

*Average 2.5-inch filter mass ± two standard deviations of 0.2159 ± 0.0030 grams.

**U-235 calculated based on natural U-235 to U-238 activity ratio of 1:21.45 (0.0466).

+ Average of the 5-filter and 6-filter composite results.

++Extrapolated value based on area ratio between 2.5-inch and 4-inch diameter filters of 0.391 times the 4-inch diameter activity content (Eadie and Bernhardt, 1976).

Table 25 presents the stack results in units of picocuries per cubic meter. These can be converted to discharge rates by multiplying by the stack flow rates given in the footnote. These values can then be related to the total plant process by multiplying by the number of similar stacks. It has been assumed that the various stacks have similar discharge parameters (i.e., there were four 200 Mill Stacks but only one such stack was sampled).

TABLE 25. STACK SAMPLING
NET RADIONUCLIDE CONCENTRATION ± TWO-SIGMA ERROR TERM, in pCi/m³

LOCATION Sampled Volume+ (Date)	Ra - 226	Po - 210**	U - 234	U - 235	U - 238	Th - 230	Th - 232	Pb - 210**	Ra - 228**
200 MILL STACK									
0.728 m³ (0/24/75)	2.9 ± 0.55	ND	ND	ND	ND	<0.0029	ND	3.6 ± 1.6	<1.3
200 PHOSPHORIC ACID SCRUBBER									
0.855 m³ (10/3/75)	0.74 ± 0.29	ND	0.064 ± 0.04	<0.01	0.10±0.042	0.18 ± 0.086	ND	3.3 ± 1.4	<1.1
TSP DRYER									
0.265 m³ (10/8/75)	9.5 ± 1.6	<4.9	8.3 ± 1.2	0.24±0.22	7.5 ±1.1	6.9 ± 1.4	<0.37	15 ± 4.7	<6.5

* ND indicates non-detectable

** Gross analytical results

+ Sampled volumes are for dry standard temperatures and pressure (STP).
The dry STP flow rates (in cubic meters per minute) for the stacks were: 200 Mill Stack-250 (four similar stacks)
200 Phosphoric Acid Scrubber-290 (three similar stack), and TSP Dryer-610 (only one stack).

It is emphasized that there are considerable uncertainties in estimating releases, especially annual releases, from single short-term samples. Such estimates are only a general indication and are impacted by variations between stacks, analytical uncertainties, daily variations in the operations of the plant, etc. Thus, without additional verification, release estimates probably have at least an order of magnitude uncertainty. Based on the reported data, the annual releases are estimated to be about 5 to 10 mCi for the uranium-238 chain nuclides (uranium-238 through lead-210). The lead-210 estimates could be somewhat higher than this, based on the previously mentioned uncertainties in the lead-210 data. The higher levels of lead-210 may be related to its volatility in the calcining and other heating processes. The low levels (compared to lead-210) of polonium-210 in the stack effluents are not readily explainable, given the volatility of polonium. It is possible a discharge point for polonium-210 was overlooked.

The stack sampling data for the 200 Mill and the 200 Phosphoric Acid Scrubber indicate somewhat higher concentrations of lead-210 and radium-226 than for the other nuclides. Uranium-234 and -238 and thorium-230 appear to be present in similar quantities as the radium-226 and lead-210 for the TSP Dryer stack sample.

The estimated stack discharges of uranium, thorium-230, radium-226, and lead-210 for the wet process plant are of the same order of magnitude (millicuries per year), but generally a bit lower than those estimated for a thermal phosphate plant (ORP/LVF, 1977). The two plants processed similar volumes of ore (1 to 2 million tons per year), but produced different products (i.e., fertilizer for the wet process plant and elemental phosphorus for the thermal process plant).

The highest stack discharges from this plant, both pCi/m^3 and pCi/sec, were for the TSP Dryer. Since this study, the air pollution control equipment for this process has been upgraded (additional cyclone and scrubber), resulting in significant decreases in the particulate and fluoride effluents and, undoubtedly an associated reduction in the quantities of radioactivity discharged to the environs.

SUMMARY

External gamma radiation measurements conducted in various working areas of the J. R. Simplot's Wet Process Plant near Pocatello, Idaho ranged from background to 120 μR/h, including an estimated background level of about 9 μR/h. Ambient radon-222 concentrations in several buildings ranged from 0.14 to a high of 1.9 pCi/l, which was measured outside the Control Room of the 300 Phosphoric Acid Plant. Out-of-doors, the ambient radon concentration ranged from 0.23 to 0.31 pCi/l in the Ore Loading and Unloading Area and the Gypsum Pile, respectively. These ambient radon concentrations are within expected ranges due to natural terrestrial background sources alone.

The highest airborne particulate radioactivity was measured in the area of the #3 Calciner Building. There the radium-226 was 1.6 pCi/m^3, and total uranium was about 2.1 pCi/m^3. These airborne concentrations are orders of magnitude greater than results obtained in the Technical Building Library.

Particle size characterization (of selected cascade impactor results) indicates geometric means of equivalent aerodynamic diameters of less than one micrometer, except for one value of six micrometers (values ranged from 0.03 to 6.0 μm). These values are within the expected ranges for atmospheric and industrial dusts. The average of all cascade impactor sampling results indicated that roughly 52 percent of the arithmetic total airborne radioactivity was contained in the particle size fraction less than one micrometer.

Difficulties in the radiochemical analysis of samples for lead-210, discovered subsequent to the completion of analysis of the reported samples, indicates up to an order of magnitude

uncertainty in many of the reported lead-210 and polonium-210 results.

Stack sampling of representative effluent discharge points showed radium-226 release concentrations ranging from 0.74 ± 0.29 to 9.5 ± 1.6 pCi/m^3. Natural uranium and lead-210 concentrations ranged up to about 15 pCi/m^3. Estimates based on these results, the number of similar stacks, and associated stack flow rates indicate annual discharges of about 5 to 10 mCi/year of lead-210, radium-226, and uranium-234 and -238, with lower values for thorium-230 and polonium-210.

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APPENDIX-A

RADIOCHEMICAL ANALYTICAL METHODS

APPENDIX-A

RADIOCHEMICAL ANALYTICAL METHODS

All analyses were completed at the Environmental Monitoring and Support Laboratory in Las Vegas, Nevada (EMSL-LV). The following sections present brief descriptions of the analytical methods employed for this study. Specific details of the procedures are contained in the Handbook of Radiochemical Analytical Methods, F. B. Johns, ed. (1975).

Analysis of Radium-226, Radium-228 and Lead-210

A sequential method for the determination of radium-226, radium-228, and lead-210 in environmental samples has been developed by the EMSL-LV. This method is initiated by the precipitation of radium from the sample aliquot using barium sulfate. Barium-radium-sulfate is then dissolved in a diethylenetriaminepentaacetate disodium solution and transferred to an emanation tube and the radon allowed to come to equilibrium, approximately 30 days ingrowth with its parent - radium. Radium-226 ($T_{\frac{1}{2}}$ = 1602 years) decays by alpha emission to radon-222 ($T_{\frac{1}{2}}$ = 3.8 days). Radon-222, a noble gas, is then collected from the liquid by a de-emanation technique. Radon-222 is usually counted for 30 minutes by alpha scintillation at four and one-half hours after the de-emanation step to allow for the build-up of the daughters.

The solution from the radium-226 determination is saved and the total radium is reprecipitated. Radium-228 ($T_{\frac{1}{2}}$ = 6.1 years) is a beta emitter and decays to actinium-228 ($T_{\frac{1}{2}}$ = 6.13 hours). The actinium is allowed to ingrow for three days and is extracted with diethylhexylphosphoric acid and back extracted with nitric acid. The actinium-228 is beta counted for 30 minutes in a low-level beta counter.

Lead is also precipitated with the radium sulfate in the original solution. Advantage is taken of the 30-day storage, for

radon-222 ingrowth, to allow the bismuth-210 to grow in. Lead-210 ($T_{\frac{1}{2}}$ = 20.4 years) decays by beta emision to bismuth-210 ($T_{\frac{1}{2}}$ = 5.01 days). The bismuth-210 is precipitated from the supernatant liquid in the radium-228 separation step. Bismuth is converted to an oxide, dissolved in nitric acid, mounted on a two-inch planchet and beta counted for 30 minutes.

As noted in the text of this report, subsequent to the analysis of the samples reported in this report, it was found that the lead-210 recovery was less than expected. This is discussed in Appendix C.

Analysis of Isotopic Uranium and Thorium

Samples are decomposed utilizing techniques of nitric-hydrofluoric acid digestion, potassium fluoride fusion or ignition. The residues are dissolved in dilute nitric acid and successive sodium and ammonium hydroxide precipitations are performed in the presence of boric acid to remove fluoride and soluble salts. The hydroxide precipitate is dissolved, the solution is adjusted to 9N in hydrochloric acid, and uranium is absorbed on an anion exchange column, separating it from thorium. Iron is removed from the column by washing with hydrochloric acid and the uranium is eluted with dilute hydrochloric acid. The thorium is converted to a nitrate form and absorbed on the same anion exchange column separating it from calcium and other interferences. The thorium is then eluted with 9N hydrochloric acid. The uranium is electrodeposited on stainless steel discs from an ammonium sulfate solution and subsequently counted by alpha spectrometry. Usually 1000-minute counting times are used for analysis. Chemical yields are normally determined by the recovery of internal tracer standards (e.g., uranium-232 and thorium-234) added at the beginning of the analysis.

Analysis of Polonium-210

Samples are decomposed by digestion with hydrofluoric acid and nitric acid in the presence of lead carrier and a polonium-208 tracer. Polonium is co-precipitated with lead sulfide from a dilute acid solution separating it from calcium, iron, and other interferences. The sulfide precipitate is dissolved in dilute hydrochloric acid and polonium is spontaneously deposited on a nickel disk. Polonium-210 and polonium-208 tracer are measured by alpha spectrometry. Usually, 1000-minute counting times are used for analysis.

Polonium-210 Activity Estimations

Due to the time delay between sample collection and radiochemical analysis for polonium-210, the following considerations have been utilized to estimate the polonium-210 activity in a sample.

1. Polonium-210 decays with a radiological half-life of 138 days. Therefore, a decay correction factor should be considered for samples which are held for a relatively long period between collection and analysis.

2. Concurrently, there will be some polonium-210 ingrowth from lead-210 and bismuth-210 contents of the original sample during the elapsed time between collection and analysis.

   The consideration of radioactive series transformations is discussed in detail in Kaplan (1958) For the specific case of lead-210, the decay scheme is as shown in Figure A-1. The solution of the system of differential equations, which describe this lead-210 decay series, was derived by Bateman and is summarized here.

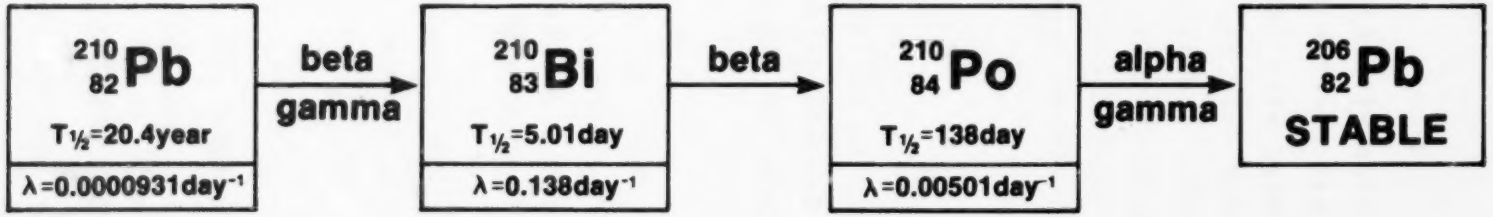

FIGURE A-1. LEAD-210 DECAY SCHEME

a. For polonium-210 ingrowth from lead-210 in the original sample:

$$A_{Po}^{Pb} =$$

$$1.01\ A_{Pb}^{o} e^{-0.0000931t} + 0.0377\ A_{Pb}^{o} e^{-0.138t} - 1.06\ A_{Pb}^{o} e^{-0.00501t}$$

Where

A_{Po}^{Pb} = polonium-210 activity at time of measurement due to ingrowth from lead-210 in sample

A_{Pb}^{o} = lead-210 activity at time of sample collection

t = Elapsed time (in days) between sample collection and analysis

Since the bismuth term is negligible and allowing for typical values of t, the following simplification of the above expression results:

$$A_{Po}^{Pb} = A_{Pb}^{o} (1.01 - 1.06\ e^{-0.00501t})$$

b. For polonium-210 ingrowth from bismuth-210 in the original sample:

$$A_{Po}^{Bi} = 0.0377\ A_{Bi}^{o} e^{-0.00501t} - 0.0377\ A_{Bi}^{o} e^{-0.138t}$$

Where

A_{Po}^{Bi} = polonium-210 activity at time of measurement due to ingrowth from bismuth-210 in sample.

A_{Bi}^{o} = bismuth-210 activity at time of sample collection

This term is insignificant for typical values of t and even in the case of the complete decay of bismuth-210, the polonium-210 ingrowth factor would be less than four percent (i.e., decay constant for polonium-210 divided by the decay constant for bismuth-210 equals 0.036).

c. The polonium-210 decay correction term:

$$A_{Po}^{Po} = A_{Po}^{o} e^{-0.00501t}$$

Where

A_{Po}^{Po} = polonium-210 activity at time of measurement due to decay of original polonium-210 in sample.

A_{Po}^{o} = Original polonium-210 activity in the sample at time of collection.

Therefore, the polonium-210 activity at the time of measurement (A_{Po}) is the summation of the two source terms (i.e. due to the ingrowth from lead-210 and due to the decay of the original polonium-210 in the sample).

$$A_{Po} = A_{Po}^{Pb} + A_{Po}^{Po}$$

or

$$A_{Po} = A_{Pb}^{o} (1.01 - 1.06\ e^{-0.00501t}) + A_{Po}^{o} e^{-0.00501t}$$

d. Due to the relatively long half-life of lead-210 ($T_{\frac{1}{2}}$ = 20.4 years), the lead-210 activity at the time of measurement (A_{Pb}) is essentially equivalent to the original Pb-210 activity in the sample (A_{Pb}^{o}). Solving the above expression for the original polonium-210 activity in the sample at the time of collection (A_{Po}^{o}) yields:

$$A_{Po}^{o} = e^{0.00501t}[A_{Po} - A_{Pb} (1.01 - 1.06\ e^{-0.00501t})]$$

All reported polonium-210 analytical results are calculated values obtained from the above equation. Whenever the apparent polonium-210 content from its ingrowth from lead-210 exceeds the measured polonium-210 activity, a non-detectable (ND) value has been reported.

e. The estimated counting error term associated with this calculated polonium-210 activity is simply the sum of squares of the individual counting error terms, or:

$$\sigma^2_{A_{Po}o} = (e^{0.005t})^2 [\sigma^2_{A_{Po}} + \sigma^2_{A_{Pb}} (1 - 1.1\, e^{-0.005t})^2]$$

Where

$\sigma_{A_{Po}o}$ = Estimated counting error term of the calculated polonium-210 activity in the sample at the time of collection.

$\sigma_{A_{Po}}$ = Counting error term of the measured polonium-210 activity.

$\sigma_{A_{Pb}}$ = Counting error term of the measured lead-210 activity.

In the tables, the estimated counting error terms at the 95 percent confidence level (i.e., twice $\sigma_{A_{Po}o}$) have been included for the calculated polonium-210 activities.

Throughout this report, the symbol for less than (<) has been used to indicate the equivalence of the error term (at the 95 percent confidence level) to the reported result.

APPENDIX-B

AIRBORNE PARTICULATE SAMPLING

GROSS RADIONUCLIDE CONCENTRATION RESULTS

TABLE B-1. AIRBORNE PARTICULATE SAMPLING
GROSS RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

RADIONUCLIDE	GRINDER MILL No. 7* 0906 to 1322 hrs. (4/30/75) pCi/m^3	pCi/g	ORE UNLOADING & STORAGE** 0945 to 1435 hrs. (5/1/75) pCi/m^3	pCi/g	GYPSUM PILE*** 0930 to 1442 hrs. (5/1/75) pCi/m^3
Ra - 226	0.23 ± 0.031	37 ± 4.9	0.16 ± 0.30	30 ± 5.6	0.10 ± 0.0045
Po - 210	0.67 ± 0.27	100 ± 43	ND	ND	0.22 ± 0.062
U - 234	0.12 ± 0.017	19 ± 2.8	0.077 ± 0.014	14 ± 2.5	0.0073 ± 0.0011
U - 235	<0.0028	<0.45	0.0084 ± 0.0045	1.5 ± 0.81	<0.00016
U - 238	0.13 ± 0.018	21 ± 2.9	0.093 ± 0.015	17 ± 2.8	0.008 ± 0.0011
Th - 230	0.29 ± 0.034	46 ± 5.4	0.17 ± 0.027	32 ± 5.0	0.032 ± 0.0026
Th - 232	0.0076 ± 0.0059	1.2 ± 0.97	0.0097 ± 0.0063	1.7 ± 1.3	0.00065 ± 0.00039
Pb - 210	0.19 ± 0.068	31 ± 11	0.24 ± 0.071	45 ± 13	0.076 ± 0.006
Ra - 228	<0.10	<16	<0.10	<19	<0.0055

*Sampled volume of 16.6m^3, with a dust particulate load of 0.1044g.

**Sampled volume of 16.6m^3, with a dust particulate load of 0.0902g.

***Sampled volume of 333m^3, collected on 8 x 10-inch of glass fiber filter.

TABLE B-2. AIRBORNE PARTICULATE SAMPLING
GROSS RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

RADIONUCLIDE	CALCINER - CONTROL ROOM* 1058 to 1520 hrs. (4/28/75) pCi/m^3	pCi/g	#3 CALCINER ** 1154 to 1335 hrs. (4/28/75) pCi/m^3	pCi/g
Ra - 226	0.073 ± 0.018	43 ± 11	1.6 ± 0.14	32 ± 2.8
Po - 210	0.60 ± 0.26	371 ± 146	9.8 ± 1.2	195 ± 24
U - 234	0.03 ± 0.0085	17 ± 5.0	1.0 ± 0.10	20 ± 2.0
U - 235	<0.0025	<1.5	0.092 ± 0.025	1.8 ± 0.50
U - 238	0.035 ± 0.0095	20 ± 5.6	1.0 ± 0.10	20 ± 2.0
Th - 230	0.063 ± 0.016	37 ± 9.5	1.8 ± 0.16	37 ± 3.1
Th - 232	<0.0037	<2.2	0.076 ± 0.038	1.5 ± 0.74
Pb - 210	0.077 ± 0.065	42 ± 36	<0.14	<2.8
Ra - 228	<0.10	<61	<0.31	<6.2

*Sampled volume of 16.5m^3, with a dust particulate load of 0.0285g.
**Sampled volume of 5.44m^3, with a dust particulate load of 0.2742g.

TABLE B-3. AIRBORNE PARTICULATE SAMPLING IN THE 300 PHOSPHORIC ACID PLANT+
GROSS RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM.

	ABOVE DIGESTER* 1016 to 1157 hrs. (4/28/75)		OUTSIDE CONTROL RM. ** 1129 to 1452 hrs. (4/28/75)		CONTINUOUS FILTER *** (LANDSKRONA) 1020 to 1233 hrs. (4/28/75)		CONTROL ROOM**** 1227 to 1537 hrs. (4/28/75)	
RADIONUCLIDE	pCi/m^3	pCi/g	pCi/m^3	pCi/g	pCi/m^3	pCi/g	pCi/m^3	pCi/g
Ra - 226	0.41 ± 0.12	27 ± 7.7	0.085 ± 0.028	13 ± 4.3	0.025 ± 0.021	8.9 ± 7.6	0.034 ± 0.017	21 ± 11
Po - 210	ND	ND	ND	ND	<0.12	<48	<0.43	<276
U - 234	0.25 ± 0.059	16 ± 3.7	0.12 ± 0.022	18 ± 3.3	0.063 ± 0.018	23 ± 6.8	0.018 ± 0.009	12 ± 6.0
U - 235	<0.023	<1.5	0.013 ± 0.0078	2.0 ± 1.2	<0.0035	<1.3	<0.0045	<3.0
U - 238	0.19 ± 0.055	12 ± 3.5	0.14 ± 0.025	21 ± 3.7	<0.0043	<1.6	0.018 ± 0.009	12 ± 6.0
Th - 230	0.44 ± 0.10	29 ± 6.5	0.22 ± 0.038	34 ± 5.9	0.074 ± 0.025	27 ± 9.1	0.027 ± 0.013	18 ± 9.0
Th - 232	0.048 ± 0.034	3.1 ± 2.3	<0.0095	<1.5	<0.0098	<3.6	<0.0045	<3.0
Pb - 210	0.82 ± 0.40	54 ± 26	0.36 ± 0.13	55 ± 19	0.18 ± 0.14	66 ± 49	0.16 ± 0.12	100 ± 78
Ra - 228	<0.49	<32	<0.17	<26	<0.19	<68	<0.17	<110

+ND indicates non-detectable
*Sampled volume of $3.02m^3$, with a dust particulate load of 0.0462g.
**Sampled volume of $10.1m^3$, with a dust particulate load of 0.0661g.
***Sampled volume of $7.91m^3$, with a dust particulate load of 0.0215g.
****Sampled volume of $8.93m^3$, with a dust particulate load 0.0134g.

TABLE B-4. AIRBORNE PARTICULATE SAMPLING+

GROSS RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM.

RADIONUCLIDE	TSP STORAGE AREA* 1020 to 1412 hrs. (4/29/75) pCi/m^3	pCi/g	TSP DRYER** 0831 to 1200 hrs. (4/30/75) pCi/m^3	pCi/g	ACIDULATION TSP DISCHARGE*** 0811 to 1207 hrs. (4/30/75) pCi/m^3	pCi/g
Ra - 226	0.019 ± 0.007	14 ± 4.9	0.57 ± 0.057	17 ± 1.7	0.017 ± 0.021	26 ± 7.8
Po - 210	<0.17	<144	1.4 ± 0.51	39 ± 15	ND	ND
U - 234	0.014 ± 0.0056	10 ± 4.1	2.1 ± 0.17	63 ± 5.1	0.068 ± 0.015	27 ± 5.9
U - 235	<0.0028	<2.0	0.054 ± 0.014	1.6 ± 0.43	0.004 ± 0.0038	1.6 ± 1.5
U - 238	0.014 ± 0.0056	10 ± 4.1	1.4 ± 0.12	41 ± 3.6	0.049 ± 0.012	19 ± 4.8
Th - 230	0.014 ± 0.0083	0 ± 6.1	2.6 ± 0.10	78 ± 3.7	0.15 ± 0.029	56 ± 11
Th - 232	<0.0028	<2.0	0.025 ± 0.012	0.73 ± 0.35	0.011 ± 0.0085	4.1 ± 3.2
Pb - 210	0.12 ± 0.079	85 ± 57	0.68 ± 0.12	20 ± 3.6	0.14 ± 0.094	53 ± 35
Ra - 228	<0.10	<84	<0.13	<3.8	0.31 ± 0.15	110 ± 55

+ND indicates non-detectable

*Sampled volume of 14.4m^3, with a dust particulate load of 0.0197g.

**Sampled volume of 11.8m^3, with a dust particulate load of 0.3998g.

***Sampled volume of 12.6m^3, with a dust particulate load of 0.0332g.

TABLE B-5. AIRBORNE PARTICULATE SAMPLING+
RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

RADIONUCLIDE	200 AMMOPHOS PLANT-DRYER* 0935 to 1201 hrs. (4/29/75) pCi/m^3	pCi/g	100 AMMOPHOS PLANT-DRYER** 0943 to 1200 hrs. (4/29/75) pCi/m^3	pCi/g
Ra - 226	0.086 ± 0.029	4.6 ± 1.6	0.098 ± 0.032	11 ± 3.7
Po - 210	ND	ND	<0.18	<20
U - 234	0.37 ± 0.048	20 ± 2.6	0.25 ± 0.036	29 ± 4.1
U - 235	0.0078 ± 0.0068	0.42 ± 0.37	0.0082 ± 0.0065	0.94 ± 0.75
U - 238	0.35 ± 0.046	18 ± 2.4	0.27 ± 0.038	31 ± 4.4
Th - 230	0.58 ± 0.066	31 ± 3.3	0.55 ± 0.072	63 ± 8.2
Th - 232	<0.0087	<0.47	<0.011	<1.3
Pb - 210	0.34 ± 0.14	18 ± 7.6	0.16 ± 0.14	18 ± 16
Ra - 228	<0.21	<11	<0.22	<25

+ND indicates non-detectable
*Sampled volume of 8.2m^3, with a dust particulate load of 0.1525g.
**Sampled volume of 7.9m^3, with a dust particulate load of 0.0686g.

TABLE B-6. AIRBORNE PARTICULATE SAMPLING+
GROSS RADIONUCLIDE CONCENTRATION ± TWO-SIGMA COUNTING ERROR TERM

RADIONUCLIDE	TECH.BLDG. LIBRARY (STORAGE)* 1036 to 1600 hrs. (4/29-30/75) pCi/m^3	pCi/g	200 AMMOPHOS PLANT** 18-46-0 STORAGE 1231 to 1600 hrs. (4/29/75) pCi/m^3	pCi/g	100 AMMOPHOS PLANT*** 11-54-0 STORAGE 1248 to 1607 hrs.(4/29/75) pCi/m^3	pCi/g
Ra - 226	<0.0012	<6.0	0.027 ± 0.014	3.2 ± 1.6	0.027 ± 0.013	6.7 ± 3.3
Po - 210	ND	ND	ND	ND	ND	ND
U - 234	0.0027 ± 0.0011	12 ± 5.2	0.24 ± 0.032	28 ± 3.8	0.15 ± 0.023	36 ± 5.5
U - 235	<0.00056	<2.5	0.0088 ± 0.0056	1.0 ± 0.67	0.0043 ± 0.004	1.0 ± 0.95
U - 238	0.0019 ± 0.0011	8.5 ± 5.0	0.24 ± 0.032	28 ± 3.7	0.16 ± 0.024	37 ± 5.6
Th - 230	0.0041 ± 0.0018	21 ± 9.1	0.48 ± 0.052	54 ± 6.1	0.28 ± 0.039	68 ± 9.4
Th - 232	<0.00038	<1.9	<0.008	<0.97	0.017 ± 0.01	4.2 ± 2.5
Pb - 210	0.048 ± 0.014	240 ± 71	0.17 ± 0.091	20 ± 11	0.14 ± 0.094	34 ± 22
Ra - 228	<0.019	<95	<0.14	<17	<0.12	<30

+ND indicates non-detectable

*Sampled volume of 19.2m^3, with a dust particulate load of 0.0179g.
**Sampled volume of 12.1m^3, with a dust particulate load of 0.1042g.
***Sampled volume of 12.3m^3, with a dust particulate load of 0.0507g.

APPENDIX C

UNCERTAINTIES IN LEAD-210 AND POLONIUM-210

AIR SAMPLE DATA

APPENDIX C

UNCERTAINTIES IN LEAD-210 AND POLONIUM-210 AIR SAMPLE DATA

Tables C-1 and C-2 present a tabulation of the polonium-210 concentrations, as of time of analysis, and lead-210 concentrations, as of the time of collection, for air samples listed in Tables 11 to 20 of the text. This tabulation can be used to assess the uncertainty associated with individual lead-210 results and to estimate the correct value.

Comparison of lead-210 interlaboratory duplicate results (ORP/ LVF, 1977) and special method evaluation samples indicates that the EMSL lead-210 results for solids and air samples may be underestimated by up to a factor of five or more. Due to the delay of about 280 days between sample collection and analysis for polonium-210, the polonium-210 values are generally indicative of the lead-210 collected in the sample. That is, while the polonium-210 collected in the sample would have decayed to 25 percent of its level at time of collection prior to analysis, the ingrowth of polonium-210 from lead-210 collected in the sample would have reached about 75 percent of the lead-210 value. Thus, the polonium-210 at time of analysis would be indicative of the lead-210 (within 50 percent) under the range of extreme conditions of a polonium-210:lead-210 ratio of one-tenth (normal ambient, NCRP, 1975) to three.

Therefore, the true lead-210 concentrations should fall between the reported lead-210 values and the polonium-210 values of time of collection (Tables C-1 and C-2). Furthermore, with minor exceptions, the lead-210 values should be near the high side of the range indicated.

The notable exceptions are cases where the polonium-210 concentrations are excessively above the reported lead-210 values (e.g., about an order of magnitude). Specifically, the sample from the #3 Calciner Area (from Table 12) indicates that the polonium-210: lead-210 ratio was probably greater than three and thus, especially for this Calciner sample, it appears the polonium-210 in this sample at time of analysis was primarily due to polonium-210 collection, and not due to ingrowth. For this sample the best estimate for lead-210 is about 0.70 pCi/m^3 or less (up to about five times the reported lead value). For the remaining air sample results, although there is some evidence of elevated polonium-210, the individual sampling results are insufficient to clearly denote even the positive presence of polonium-210 (beyond that ingrown from the lead-210). That is, a polonium-210:lead-210 ratio of up to four at time of analysis for these data (if analysis is at 280 days after collection) can be solely due to a factor of five error in the lead-210 analysis and 75 percent ingrowth of polonium-210 from the lead. Although such a situation of elevated lead-210 with no polonium-210 is not likely, much of the data are insufficient to demonstrate that it could not occur.

In summary, the best estimates for the lead-210 values are the polonium-210 values at time of analysis, except for the value denoted above. The polonium-210 values can only be approximated as discussed above.

TABLE C-1. ESTIMATED UNCERTAINTY FOR LEAD-210 IN AIR FILTERS*+

Referenced Table/ Location Description	Reported Lead-210 (pCi/m^3)	Polonium-210 at Time of Analysis (pCi/m^3)
Table 11		
Grinder Mill No. 7	0.19 ± 0.068	0.30 ± 0.041
Ore Unloading and Storage	0.24 ± 0.071	0.11 ± 0.033
Gypsum Pile	0.076 ± 0.006	0.11 ± 0.015
Table 12 - Calciner Area		
Control Room	0.077 ± 0.065	0.20 ± 0.04
#3 Calciner	<0.14	2.5 ± 0.27
Table 13 - 300 Phosphoric Plant		
Above Digester	0.82 ± 0.40	0.53 ± 0.13
Outside Control Room	0.36 ± 0.13	0.24 ± 0.047
Continuous Filter	0.18 ± 0.14	0.16 ± 0.052
Control Room	0.16 ± 0.12	0.22 ± 0.088
Table 14		
TSP Storage Area	0.12 ± 0.079	0.13 ± 0.029
TSP Dryer	0.68 ± 0.12	0.83 ± 0.084
Acidulation TSP Discharge	0.14 ± 0.094	0.094 ± 0.026
Table 15		
200 Ammophos Plant - Dryer	0.34 ± 0.014	0.15 ± 0.068
100 Ammophos Plant - Dryer	0.16 ± 0.14	0.16 ± 0.14
Table 16		
Library	0.048 ± 0.014	0.025 ± 0.0063
200 Ammophos Plant-Storage	0.17 ± 0.091	0.12 ± 0.0063
100 Ammophos Plant-Storage	0.14 ± 0.094	0.093 ± 0.056

* These data are gross results prior to blank filter activity subtraction.

\+ The statistical uncertainty of values as expressed by the two-sigma counting error must be considered in comparing values. For example, 5 ± 2 is not statistically greater than 3 ± 1. Furthermore, counting error does not include other analytical and sample collection uncertainties.

TABLE C-2. ESTIMATED UNCERTAINTY FOR LEAD-210 IN AIR FILTERS*+

Referenced Table/ Location Description	Reported Lead-210 (pCi/m^3)	Polonium-210 at Time of Analysis (pCi/m^3)
Table 17. Ore Unloading Area		
Filter 1	0.07 ± 0.035	0.09 ± 0.024
2	0.074 ± 0.035	0.039 ± 0.014
3	0.088 ± 0.036	0.056 ± 0.018
4	0.081 ± 0.036	0.042 ± 0.009
5	0.083 ± 0.036	0.044 ± 0.027
Final filter	0.21 ± 0.043	0.25 ± 0.028
Table 18. Calciner Building		
Filter 1	0.088 ± 0.033	0.13 ± 0.028
2	0.023 ± 0.019	0.077 ± 0.015
3	<0.03	0.19 ± 0.094
4	<0.032	0.082 ± 0.02
5	0.050 ± 0.032	0.10 ± 0.031
Final filter	0.12 ± 0.036	0.27 ± 0.03
Table 19. 200 Ammophos Plant		
Filter 1	0.11 ± 0.037	0.096 ± 0.024
2	0.073 ± 0.035	0.083 ± 0.024
3	0.10 ± 0.037	0.067 ± 0.018
4	0.07 ± 0.036	0.04 ± 0.016
5	0.097 ± 0.037	0.052 ± 0.016
Final filter	<0.032	0.052 ± 0.014
Table 20. Gypsum Pile		
Filter 1	0.011 ± 0.0051	0.007 ± 0.0025
2	0.011 ± 0.0052	0.0077± 0.0026
3	0.017 ± 0.0055	0.008 ± 0.0022
4	0.015 ± 0.005	0.007 ± 0.002
5	0.017 ± 0.0051	0.012 ± 0.0025
Final filter	0.11 ± 0.016	0.13 ± 0.014

* These data are gross results prior to blank filter activity subtraction.

\+ The statistical uncertainty of values as expressed by the two-sigma counting error must be considered in comparing values. For example, 5 ± 2 is not statistically greater than 3 ± 1. Furthermore, counting error does not include other analytical and sample collection uncertainties.